Averaging Rules for the Scattering by Randomly Oriented Chiral Particles

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Abstract— **The orientational averages arising when calculating the effective behavior of ensembles of randomly dispersed (chiral) particles are examined. Two methods, i.e., the vector radiative transfer equation (VRTE) and the Lorenz–Lorentz mixing formulas, are briefly discussed. Since integration over products of up to six elements of a rotational matrix are involved, the effort for performing the averaging is high. To minimize the computational burden, rules for evaluating the integrals will be given in this communication. Application to average polarizability-tensors are presented.**

I. Introduction

For calculating the effective electric properties of a chiral medium from the knowledge of the behavior of a single element, several methods have been applied in the past. Among them one finds the use of the chiral Lorenz–Lorentz formulas [1]–[4] as developed by Sihvola and Lindell [5], scattering approaches based on the vector radiative transfer equation (VRTE) [3], or purely numerical techniques [6]. In all of these methods averaging procedures are needed in order to extract the effective parameters of the random medium under consideration.

Applying the Lorenz–Lorentz formulas, where the ensemble is approximated as a homogeneous medium, requires the chiral particles to be modeled as small dipoles. In general, the underlying polarizabilities of nonspherical particles are tensor-quantities that describe the electric, magnetic, and magnetoelectric (cross)coupling [7]. For randomly oriented and distributed particles, these need to be averaged in order to calculate the effective behavior of the ensemble.

The VRTE, based on the concept of generalized Stokes parameters, was developed to describe the effective homogeneous and inhomogeneous scattering of sparse ensembles of small particles [8]. The coefficients of the VRTE, i.e., the so-called extinction and phase matrices, can be calculated from the scattering matrix of a single particle, which follows directly from the polarizabilities [3].

In both cases, averages with respect to the different orientations of the scatterers that are described by the Eulerian matrix need to be evaluated. To obtain the extinction and the phase matrix for ensembles of chiral scatterers up to third- and sixth-order combinations of elements of the rotation matrix have to be averaged, respectively, whereas the average polarizabilities involve up to third-order combinations.

In this paper we will indicate the rules for up-to-sixth-order combinations. The extinction and phase matrices can then be calculated by employing the rules developed here [3], [8]. Here, the application of these rules will be demonstrated with two simple examples, namely the calculation of average polarizabilities.

To our knowledge this is the first comprehensive publication of the development of these rules.

II. Polarizabilities of Arbritrarily Oriented Elements

Two coordinate systems need to be considered. The incident wave is defined with respect to the lab-system, indicated by a prime ($'$), whereas a single particle is described in a local system. The two systems can be transformed into each other by means of the Eulerian matrix $\overline{\overline{A}}$ [8], which performs a change of basis and thus allows to calculate the scattering properties of a particle with a given orientation from those of an identical, but differently oriented, element.

Within the well-known dipole approximation, valid for particles much smaller than the wavelength in the respective far-fields, the electric behavior of a single particle can be expressed by electric, magnetic and magnetoelectric (cross-)polarizabilities. In the local coordinate system the dipole moments of a single, small chiral element is given by

$$\vec{p} = [\overline{\overline{p}}_{ee} + \overline{\overline{p}}_{em}(\hat{k}^{inc})]\,\vec{E}^{inc}, \tag{1}$$

$$\vec{m} = [\overline{\overline{p}}_{me} + \overline{\overline{p}}_{mm}(\hat{k}^{inc})]\,\vec{E}^{inc} \tag{2}$$

where $\vec{p}$ and $\vec{m}$ are the electric and magnetic dipole moments and $\overline{\overline{p}}_{ij}$ denote the polarizability tensors. The first subscript refers to the effect and the second to the origin of the moment. Here, both the electric and the magnetic excitation are expressed by the incident electric field $\vec{E}^{inc}$ and, implicitly, the direction of incidence $\hat{k}^{inc}$.

Once the polarizabilities of a scatterer are known for a given angle of incidence of the incoming wave, those of tilted but otherwise identical elements directly follow from them and from the knowledge of the rotation relative to the excitation. Mathematically, this corresponds to a rotation of the local coordinate system with respect to a reference orientation, which is defined in the lab coordinate system. Thus, the polarizabilities of tilted scatterers can be obtained from

$$\overline{\overline{p}}'_{ie} = \overline{\overline{A}}\,\overline{\overline{p}}_{ie}\,\overline{\overline{A}}^{T} \tag{3}$$

when the electric field is the origin. Since the magnetically induced polarizations depend on the propagation direction of the incident wave, which is fixed with respect to the lab system, the transformations become more complicated in this case

$$\overline{\overline{p}}'_{im} = \overline{\overline{A}}\left[\frac{\vec{\overline{\overline{\alpha}}}_{im}}{\eta}\left(\overline{\overline{A}}^{T}\,\hat{k}'_{inc}\right)\right]\overline{\overline{A}}^{T}. \tag{4}$$

Here, $\vec{\overline{\overline{\alpha}}}_{im}$ follows from the usual polarizability via

$$\overline{\overline{p}}_{im}(\hat{k}^{inc}) = \frac{\vec{\overline{\overline{\alpha}}}_{im}}{\eta}\cdot\hat{k}^{inc} \tag{5}$$

where the matrix vector $\vec{\overline{\overline{\alpha}}}_{im}$ has been introduced. It is a three-dimensional vector with 3×3 matrices as elements. The dot-product with a vector is then a multiplication of each element with the respective submatrix. The notation of the scalar element $\alpha_{im(jkl)}$ is as follows: j denotes the vector element, whereas kl is the usual matrix notation within the submatrix.

III. Orientational Averaging

Now, the average polarizabilities can be determined. Averaging over all possible orientations of an orientation-dependent matrix $\overline{\overline{q}}$ requires the integral

$$\langle\overline{\overline{q}}\rangle = \frac{\int_0^{2\pi}\int_0^{\pi}\int_0^{2\pi}\overline{\overline{q}}(\alpha,\beta,\gamma)f(\alpha,\beta,\gamma)\sin\beta\,d\alpha\,d\beta\,d\gamma}{\int_0^{2\pi}\int_0^{\pi}\int_0^{2\pi}f(\alpha,\beta,\gamma)\sin\beta\,d\alpha\,d\beta\,d\gamma} \tag{6}$$

to be solved.

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Here, α, β, and γ describe the angles of rotation between the lab and the local system, while $f(\alpha, \beta, \gamma)$ is the probability density function for the orientations. In the following it is set to unity, assuming a uniform distribution of all orientations. When applying (6) to the polarizabilities, it is clear from (3) and (4) that products of at most three elements of the Eulerian matrix need to be averaged. The extinction and the phase matrices of the VRTE, in turn, would involve products of up to six elements.

Even though the integrals arising from (6) can in general be calculated numerically, this cannot be done economically for the arising high-order products. Therefore, we shall find vanishing integrals analytically and eliminate them from the calculation. The remaining combinations can easily be calculated and tabulated in rules.

The Eulerian matrix is defined by

$$\overline{\overline{A}} = \{a_{ij}\}_{i,j=1,2,3}. \tag{7}$$

Here a_{ij} is a short-hand notation for the dot product $\hat{a}_i \cdot \hat{b}_j$, where $\hat{a}_i$ and $\hat{b}_j$ $(i, j = 1, 2, 3)$ are the basis vectors of the local and the lab system, respectively.

Depending on their order, the combinations of these elements will lead to different rules that will be derived next. For the logical operations we have used the following symbols: $\wedge$ stands for AND, $\vee$ for OR, and $\oplus$ for XOR. We shall start with order two and observe that the identity

$$a_{ij}a_{kl} = \tfrac{1}{2}\left[\cos(\lambda+\mu) + \cos(\lambda-\mu)\right] \tag{8}$$

holds. Here, λ and μ are the angles between the vectors of each considered pair

$$a_{ij} = \cos(\lambda) \tag{9}$$

$$a_{kl} = \cos(\mu). \tag{10}$$

They are determined by the relative orientations α, β, and γ of the two coordinate systems, and therefore a_{ij}, a_{kl} can be expressed in terms of these angles. It can then be shown that the integral (6) will vanish if $\lambda \neq \mu$, that is $i \neq k \vee j \neq l$. In all other cases the integral can be calculated and is found to be $\frac{1}{3}$. This result can be expressed as the first rule.

Rule 2:

$$\langle a_{ij}a_{kl}\rangle = \begin{cases} \frac{1}{3} & \text{if } i = k \wedge j = l, \\ 0 & \text{otherwise.} \end{cases} \tag{11}$$

In order to find the average of three elements of the Eulerian matrix we can write

$$a_{ij}a_{kl}a_{mn} = \tfrac{1}{4}\left[\cos(\lambda+\mu+\nu) + \cos(\lambda+\mu-\nu) + \cos(\lambda-\mu+\nu) + \cos(-\lambda+\mu-\nu)\right] \tag{12}$$

where, again, λ, μ, and ν are the angles between the vectors of the respective pairs. The mean value of this expression does not vanish only if one of the cosine terms is constant for all angles α, β, and γ. This is the case if all the dot products involve mutually different basis vectors of the systems. Notice there are never two constant terms within the same constellation. Computing the nonzero terms yields

Rule 3:

$$\langle a_{ij}a_{kl}a_{mn}\rangle = \begin{cases} \frac{1}{6} & i \neq k \neq m \wedge j \neq l \neq n \\ & \wedge\, j,\, l,\, n \text{ increasing}, \\ -\frac{1}{6} & i \neq k \neq m \wedge j \neq l \neq n \\ & \wedge\, j,\, l,\, n \text{ decreasing}, \\ 0 & \text{otherwise} \end{cases} \tag{13}$$

where $i < k < m$ holds. As can be seen, the sense (left or right) of the connection between the two coordinate systems is important.

The following higher order combinations can always be reduced to the two basic rules developed above. This leads to the following.

Rule 4:

$$\begin{aligned} \langle a_{ij}^4\rangle &= \tfrac{1}{5}, \\ \langle a_{ij}^2 a_{kl}^2\rangle &= \begin{cases} \frac{1}{15} & i = k \oplus j = l, \\ \frac{2}{15} & i \neq k \wedge j \neq l, \end{cases} \\ \langle a_{ij}a_{kl}a_{mn}a_{op}\rangle &= 0 \text{ in the remaining cases.} \end{aligned} \tag{14}$$

Rule 5:

$$\begin{aligned} \langle a_{ij}^5\rangle &= 0, \\ \langle a_{ij}^4 a_{kl}\rangle &= 0, \\ \langle a_{ij}^3 a_{kl}a_{mn}\rangle &= \tfrac{3}{5}\langle a_{ij}a_{kl}a_{mn}\rangle \\ &\qquad i \neq k,\, m \vee j \neq l,\, n, \\ \langle a_{ij}^2 a_{kl}a_{mn}a_{op}\rangle &= \tfrac{1}{5}\langle a_{kl}a_{mn}a_{op}\rangle \\ &\qquad i \neq k,\, m,\, o \vee j \neq l,\, n,\, p, \\ \langle a_{ij}a_{kl}a_{mn}a_{op}a_{qr}\rangle &= 0 \text{ in the remaining cases.} \end{aligned} \tag{15}$$

In the third and fourth cases Rule 3 applies.

Rule 6:

$$\begin{aligned} \langle a_{ij}^6\rangle &= \tfrac{1}{7}, \\ \langle a_{ij}^4 a_{kl}^2\rangle &= \begin{cases} \frac{1}{35} & i = k \oplus j = l, \\ \frac{3}{35} & i \neq k \wedge j \neq l, \end{cases} \\ \langle a_{ij}^2 a_{kl}^2 a_{mn}^2\rangle &= \begin{cases} \frac{1}{105} & (1), \\ \frac{2}{105} & (2), \\ \frac{1}{35} & (3), \\ \frac{8}{105} & (4), \end{cases} \\ \langle a_{ij}^2 a_{kl}a_{mn}a_{op}a_{qr}\rangle &= \tfrac{3}{7}\sum_{S_1}\left[\langle a_{ij}a_b a_c\rangle \times \langle a_{ij}a_d a_e\rangle\right], \\ \langle a_{ij}a_{kl}a_{mn}a_{op}a_{qr}a_{st}\rangle &= \tfrac{12}{35}\sum_{S_2}\left[\langle a_b a_c a_d\rangle \times \langle a_e a_f a_g\rangle\right], \\ \langle a_{ij}a_{kl}a_{mn}a_{op}a_{qr}a_{st}\rangle &= 0 \text{ in the remaining cases.} \end{aligned} \tag{16}$$

The numbers above denote:

1) All the subscripts in one position are equal: $(i = k = m \wedge j \neq l \neq n) \oplus (i \neq k \neq m \wedge j = l = n)$. Thus, the expression is of the form $a_{ij}^2 a_{il}^2 a_{in}^2$ or $a_{ij}^2 a_{kj}^2 a_{mj}^2$.
2) The combinations have pairwise one subscript at the same position in common, and are expressions of the form $a_{ij}^2 a_{il}^2 a_{kl}^2$, $i \neq k \wedge j \neq l$.
3) Only one pair of the combinations has one subscript at the same position in common, and the expressions are, for instance, of the form $a_{ij}^2 a_{il}^2 a_{mn}^2$, $i \neq m \wedge j \neq l \neq n$.
4) All subscripts at the same position are unequal, and the expressions are of the form $a_{ij}^2 a_{kl}^2 a_{mn}^2$ with $i \neq k \neq m \wedge j \neq l \neq n$.

In the fourth and fifth cases we only consider pairwise unequal subscript doublets. The other combinations either are included in the previous cases or will lead to zero (in the last case). The sets S_1 and S_2 are the ordered permutations of the subscript pairs, the parenthesis $(\cdots)$ denoting ordered sets:

- For the fourth case let

$$\begin{aligned} M &= \{([kl], [mn], [op], [qr])\}, \\ \text{then} \quad S_1 &= \{[(b, c), (d, e)] \mid b, c \in M \\ &\qquad \wedge\, d, e \in M \setminus \{b, c\} \wedge b \neq c \wedge d \neq e\}. \end{aligned} \tag{17}$$

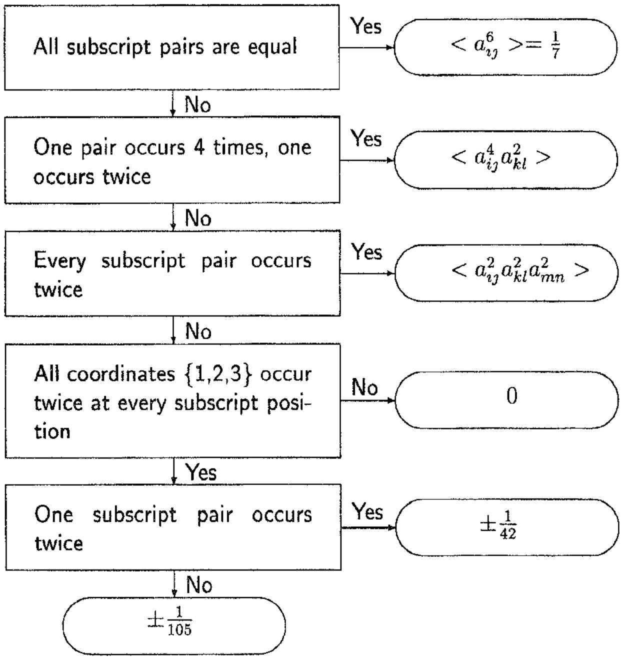

Fig. 1. Decision tree for Rule 6.

- For the fifth case let

$$M = \{([ij], [kl], [mn], [op], [qr], [st])\},$$
$$\text{then } S_2 = \{[(b, c, d), (e, f, g)] | b, c, d \in M \wedge e, f, g \in M \setminus \{b, c, d\} \wedge b \neq c \neq d \wedge e \neq f \neq g\}. \quad (18)$$

Again, in these cases the averages require the use of Rule 3, which also determines the sign.

Rule 6 can be expressed more easily in a decision tree as shown in Fig. 1.

IV. Application to the Polarizabilities

The rules for averaging the elements of the Eulerian matrix in combinations of different orders have been stated above. Let us now examine two simple cases and use the results to average the polarizability tensors. From (3) we obtain

$$\langle \overline{\overline{p}}_{ie} \rangle = \tfrac{1}{3}(p_{ie11} + p_{ie22} + p_{ie33}). \quad (19)$$

This is the well-known result for the orientational average of a tensor of rank two with respect to a uniform distribution of orientations. For the polarizability with magnetic origin Rule 3 applies and leads together with (4) to

$$\langle \overline{\overline{p}}_{im} \rangle \vec{E}^{inc} = \frac{1}{6\eta}[\alpha_{im(123)} - \alpha_{im(132)} - \alpha_{im(213)} + \alpha_{im(231)} + \alpha_{im(312)} - \alpha_{im(321)}] \cdot \vec{k}^{inc} \times \vec{E}^{inc}. \quad (20)$$

This shows that the magnetic polarizabilities basically reveal the structure of a vector product with the direction of the incident field, as expected. Together with the Lorenz–Lorentz formulas for chiral media [5], it can be employed to calculate the effective material parameters of a chiral medium that consists of chiral elements randomly dispersed in a nonchiral host medium [1]–[4].

V. Conclusion

Rules for orientational averages as they are encountered in calculations of effective material parameters with, e.g., mixing formulas or the vector radiative transfer equation have been presented. Since for the (cross-)polarizabilities with magnetic origin higher-order combinations of the Eulerian matrix elements are involved, the averaging integral in (6) cannot be solved efficiently by numerical or straight-forward analytical methods. Fast solutions for up to sixth-order combinations have been formulated in form of rules. Applying these significantly reduces computing time and memory space when calculating the average behavior of chiral particles. The cases shown as examples are important, when the effective parameters of a chiral medium are calculated from the Lorenz–Lorentz formulas. Of course, the rules presented here also apply to nonchiral scatterers.

Note: Just as our paper was going to press, we became aware of [9] and [10]. Several results obtained by us also appear in these papers, although in a different context, as well as in notation largely unfamiliar to the microwave community.

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