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Stokes parameters for light scattering from a Faraday-active sphere

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Abstract

We present an exact calculation for the scattering of light from a single sphere made of Faraday-active material, to first order in the external magnetic field. We use a recent expression for the T-matrix of a Mie scatterer in a magnetic field to compute the Stokes parameters in single scattering that describe flux and polarization of the scattered light. © 1999 Elsevier Science Ltd. All rights reserved.

1. Introduction

Several reasons exist why one wishes to understand light scattering from a dielectric sphere made of magneto-active material. Single scattering is the building block for multiple scattering. Recently, many experiments such as those reported by Erbacher et al. [1] and Rikken et al. [2] have been done with diffuse light in a magnetic field. It turns out that the theory using point-like scatterers in a magnetic field, as first developed by MacKintosh and John [3], does not always enable a quantitative analysis, for the evident reason that experiments do not contain "small" scatterers. This paper addresses light scattering from a sphere of any size in a homogeneous magnetic field.

The model of Rayleigh scatterers was used successfully to describe specific properties of multiple light scattering in magnetic fields, such as for instance *Coherent Backscattering*, *Photonic Hall Effect* (PHE) and *Photonic Magneto-resistance* (PMR). The first study of one gyrotropic sphere, due to Ford and Werner [4], was applied to the scattering of semiconducting spheres by Dixon and Furdyna [5]. For the case of magneto-active particles, for which the change in the dielectric

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constant induced by the magnetic field is small, a perturbational approach is in fact sufficient. Kuzmin [6] showed that the problem of scattering by a weakly anisotropic particle of any type of anisotropy can be solved to first order in the perturbation. Using a T-matrix formalism, Lacoste et al. [7] developed independently a perturbational approach for the specific case of magneto-optical anisotropy. This was successfully applied to compute the diffusion coefficient for magneto-transverse light diffusion [8]. Using the T-matrix for a Mie scatterer in a magnetic field we have obtained, we discuss the consequences for the Stokes parameters [9] that describe the polarization of the scattered light.

2. Perturbation theory

In this paper we set $c_0 = 1$. In a magnetic field $\mathbf{B}$, the refractive index is a tensor of rank two. For the standard Mie problem, its value at position $\mathbf{r}$ depends on the distance to the center of the sphere $|\mathbf{r}|$, which has a radius a via the Heaviside function $\Theta(|\mathbf{r}| - a)$, that equals 1 inside the sphere and 0 outside,

$$\varepsilon(\mathbf{B}, \mathbf{r}) - \mathbf{I} = [(\varepsilon_0 - 1)\mathbf{I} + \varepsilon_F \mathbf{\Phi}]\Theta(|\mathbf{r}| - a). \tag{1}$$

In this expression, $\mathbf{I}$ is the identity tensor, $\varepsilon_0 = m^2$ is the value of the normal isotropic dielectric constant of the sphere of relative index of refraction m (which is allowed to be complex-valued) and $\varepsilon_F = 2mV_0B/\omega$ is a dimensionless coupling parameter associated with the amplitude of the Faraday effect (V_0 being the Verdet constant, B the amplitude of the magnetic field, and ω the frequency). We introduced the antisymmetric hermitian tensor $\Phi_{ij} = i\varepsilon_{ijk}\hat{B}_k$ (the hat above vectors notes normalized vectors). The Mie solution depends on the dimensionless size parameters $x = \omega a$ and $y = mx$. In this paper we restrict ourselves to non-absorbing media so that m and ε_F are real-valued. Since $\varepsilon_F \approx 10^{-4}$ in most experiments, a perturbational approach is valid.

Upon noting that the Helmholtz equation is formally analogous to a Schrödinger equation with potential $\mathbf{V}(\mathbf{r}, \omega) = [\mathbf{I} - \varepsilon(\mathbf{B}, \mathbf{r})]\,\omega^2$ and energy ω^2, the T-operator is given by the following Born series:

$$\mathbf{T}(\mathbf{B}, \mathbf{r}, \omega) = \mathbf{V}(\mathbf{r}, \omega) + \mathbf{V}(\mathbf{r}, \omega)\cdot \mathbf{G}_0(\omega, \mathbf{p})\cdot \mathbf{V}(\mathbf{r}, \omega) + \cdots . \tag{2}$$

Here $\mathbf{G}_0(\omega, \mathbf{p}) = 1/(\omega^2\mathbf{I} - p^2\mathbf{\Delta}_p)$ is the free Helmholtz Green's operator in Gaussian rationalized units for pure dielectric particles, and $\mathbf{p} = -i\nabla$ is the momentum operator. The tensor of rank two $(\Delta_p)_{ij} = \delta_{ij} - p_ip_j/p^2$ projects upon the space transverse to the direction of $\mathbf{p}$. The T-matrix is defined as

$$\mathbf{T}_{\mathbf{k}\sigma,\mathbf{k}'\sigma'} = \langle \mathbf{k}, \sigma|\mathbf{T}\,|\,\mathbf{k}', \sigma'\rangle, \tag{3}$$

where $|\,\mathbf{k},\sigma\rangle$(respectively, $|\,\mathbf{k}',\sigma'\rangle$) represents an incident (respectively, emergent) plane wave with direction $\mathbf{k}$ and state of helicity σ (respectively, $\mathbf{k}'$ and σ').We will call $\mathbf{T}^0$ the part of $\mathbf{T}$ that is independent of the magnetic field and $\mathbf{T}^1$ the part of the T-matrix linear in $\mathbf{B}$. We have found the following result [7]:

$$\mathbf{T}^1_{\mathbf{k}\sigma,\mathbf{k}'\sigma'} = \varepsilon_F\,\omega^2\langle \Psi^-_{\mathbf{k},\sigma}|\Theta\,\mathbf{\Phi}|\Psi^+_{\mathbf{k}',\sigma'}\rangle, \tag{4}$$

where the $\Psi^{\pm}_{\mathbf{k},\sigma}(\mathbf{r})$ are the unperturbed eigenfunctions of the conventional Mie problem. This eigenfunction represents the electric field at the point $\mathbf{r}$ for an incident plane wave $|\mathbf{k},\sigma\rangle$. This eigenfunction is "outgoing" for $\Psi^{+}_{\mathbf{k},\sigma}$ and "ingoing" for $\Psi^{-}_{\mathbf{k},\sigma}$. Eq. (4) resembles the perturbation formula for the Zeeman shift in terms of the atomic eigenfunctions, although here it provides a complex-valued amplitude in terms of *continuum* eigenfunctions, rather than a real-valued energy shift in terms of bound states.

3. *T* matrix for Mie scattering

In order to separate radial and an angular contribution in Eq. (4), we used a well-known expansion of the Mie eigenfunction $\Psi^{+}_{\mathbf{k},\sigma}$ in the basis of vector spherical harmonics [10]. We choose the quantification axis z along the magnetic field. With this choice, the operator $\mathbf{S}_z$, the z-component of a spin one operator, can be associated with the tensor $-\mathbf{\Phi}$. The eigenfunctions of the operator $\mathbf{S}_z$ form a convenient basis for the problem. The expansion of Eq. (4) in vector spherical harmonics leads to a summation over quantum numbers J, J', M and M'. The Wigner–Eckart theorem applied to the vector operator $\mathbf{S}$ gives the selection rules for this case $J = J'$ and $M = M'$.

The radial integration can be done using a method developed by Bott et al. [11], which gives,

$$\mathbf{T}^1_{\mathbf{k},\mathbf{k}'} = \frac{16\pi}{\omega} W \sum_{J,M} (-M)\left[\mathscr{C}_J \mathbf{Y}^e_{J,M}(\hat{\mathbf{k}})\mathbf{Y}^{e*}_{J,M}(\hat{\mathbf{k}}') + \mathscr{D}_J \mathbf{Y}^m_{J,M}(\hat{\mathbf{k}})\mathbf{Y}^{m*}_{J,M}(\hat{\mathbf{k}}')\right] \tag{5}$$

with the dimensionless parameter:

$$W = V_0 B \lambda.$$

λ is the wavelength in the medium. The meaning of the indices e, m is explained in Appendix A. In the limiting case of a perfect dielectric sphere with no absorption ($\Im m(m) \to 0$), the coefficients are given by

$$\mathscr{C}_J = \frac{-c_J^{2*} u_J^2 y}{J(J+1)}\left(\frac{A_J}{y} - \frac{J(J+1)}{y^2} + 1 + A_J^2\right), \tag{6}$$

$$\mathscr{D}_J = \frac{-d_J^{2*} u_J^2 y}{J(J+1)}\left(-\frac{A_J}{y} - \frac{J(J+1)}{y^2} + 1 + A_J^2\right) \tag{7}$$

with $A_J(y) = u'_J(y)/u_J(y)$, $u_J(y)$ the Ricatti–Bessel function, and c_J and d_J the Mie amplitude coefficients of the internal field [9].

Two important symmetry relations must be obeyed by our T-matrix. The first one is parity symmetry and the second one reciprocity. These relations can be established generally when the Hamiltonian of a given system has the required symmetries (cf. Eq. (15.53) and Eq. (15.59a) P454 of Ref. [10]). We give in Appendix A a less general derivation of these relations for our specific problem:

$$T_{-\mathbf{k}\sigma,-\mathbf{k}'\sigma'}(\mathbf{B}) = T_{\mathbf{k}\sigma,\mathbf{k}'\sigma'}(\mathbf{B}), \tag{8}$$

$$T_{-\mathbf{k}'-\sigma',-\mathbf{k}-\sigma}(-\mathbf{B}) = T_{\mathbf{k}\sigma,\mathbf{k}'\sigma'}(\mathbf{B}). \tag{9}$$

We emphasize that $\sigma(-\mathbf{k}) = -\sigma(\mathbf{k})$, i.e. σ indicates in fact helicity and *not* circular polarization. The helicity is the eigenvalue of the operator $\mathbf{S}\cdot\mathbf{k}$.

3.1. The amplitude matrix

The amplitude matrix $\mathbf{A}$ relates incident and scattered field with respect to an arbitrary plane of reference. A common choice is the plane that contains the incident and the scattered wave vector, and which is for this reason called the scattering plane. We will call the linear base the base made of one vector in this plane and one vector perpendicular to it. In this basis, the amplitude matrix sufficiently far $\omega r \gg 1$, is simply defined from the T-matrix by

$$\mathbf{A}_{\mathbf{k},\mathbf{k}'} = \frac{-1}{4\pi r}\,\mathbf{T}^*_{\mathbf{k},\mathbf{k}'} = \frac{e^{i\phi}}{i\omega r}\begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix}. \tag{10}$$

ϕ is a phase factor that depends on the relative phase of the scattered wave with respect to the incident wave and which is defined in Ref. [9]. The complex conjugation in Eq. (10) is simply due to a different sign convention in Newton [10]. When no magnetic field is applied, the T-matrix of the conventional Mie-problem is given by a formula analogous to Eq. (5) where $\mathscr{C}_J$ and $\mathscr{D}_J$ have been replaced by the Mie coefficients a_J and b_J, and with $M = 1$. Because of rotational invariance of the scatterer, the final result only depends on $\cos\theta$, the scalar product of $\mathbf{k}$ and $\mathbf{k}'$, θ is the scattering angle. Therefore, we get in the circular basis (associated with the helicities σ and σ'):

$$T^0_{\sigma\sigma'} = \frac{2\pi}{i\omega}\sum_{J\geq 1}\frac{2J+1}{J(J+1)}\,(a_J^* + \sigma\sigma' b_J^*)[\pi_{J,1}(\cos\theta) + \sigma\sigma'\tau_{J,1}(\cos\theta)]. \tag{11}$$

Alternatively, the T-matrix may be expanded on the basis of the Pauli matrices

$$\mathbf{T}^0 = \frac{2\pi}{i\omega}\,[(S_1^* + S_2^*)\mathbf{I} + (S_1^* - S_2^*)\sigma_x]. \tag{12}$$

In Eq. (11), the polynomials $\pi_{J,M}$ and $\tau_{J,M}$ are defined in terms of the Legendre polynomials P_J^M by [9],

$$\pi_{J,M}(\theta) = \frac{M}{\sin\theta}\,P_J^M(\cos\theta), \qquad \tau_{J,M}(\theta) = \frac{d}{d\theta}\,P_J^M(\cos\theta). \tag{13}$$

For $M = 1$, $\pi_{J,1}$ and $\tau_{J,1}$ are polynomials of $\cos\theta$ of order $J-1$ and J, respectively, but not in general for any value of M. When written in the linear basis of polarization, Eq. (11) implies that a Mie scatterer has $S_3 = S_4 = 0$ as imposed by the rotational symmetry. For the backward direction $\theta = \pi$, the reciprocity symmetry implies that $S_3 + S_4 = 0$ for an arbitrary particle (possibly non-spherical) [9]. We will see that these two properties do not hold anymore when a magnetic field is present.

3.2. General case for $\mathbf{T}^1$ when $\hat{\mathbf{k}}\times\hat{\mathbf{k}}' \neq 0$

It remains to express the vector spherical harmonics in Eq. (5), as a function of the natural angles of the problem. In Fig. 1, we give a schematic view of the geometry. In the presence of a magnetic

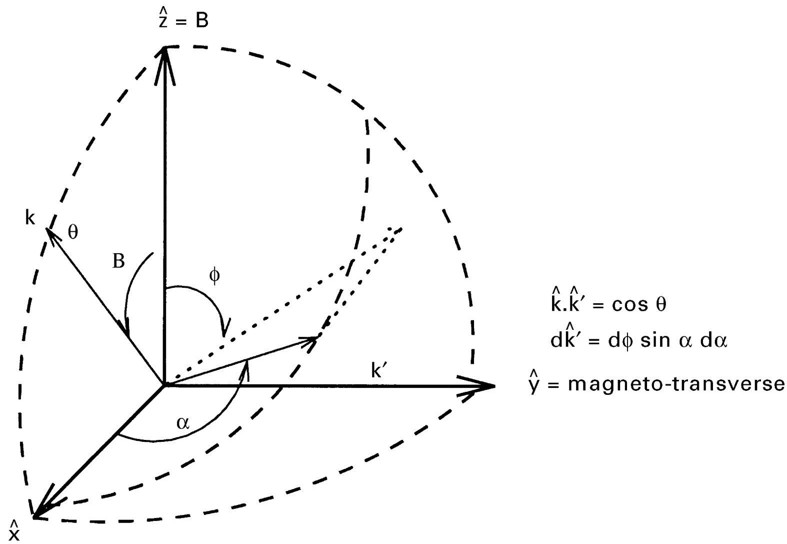

Fig. 1. Schematic view of the magneto-scattering geometry. Generally, θ denotes the angle between incident and outgoing wave vectors, ϕ is the azimuthal angle in the plane of the magnetic field and the y-axis. This latter is by construction the magneto-transverse direction defined as the direction perpendicular to both the magnetic field and the incident wave vector. Angle α coincides with angle θ in the special but relevant case that the incident vector is normal to the magnetic field.

field, the rotational invariance is broken because $\mathbf{B}$ is fixed in space. Because our theory treats $\mathbf{T}^1$ linear in $\hat{\mathbf{B}}$, $\mathbf{T}^1$ can be constructed by considering only three special cases for the direction of $\hat{\mathbf{B}}$. If $\hat{\mathbf{k}}$ and $\hat{\mathbf{k}}'$ are not collinear, we can decompose the unit vector $\hat{\mathbf{B}}$ in the non-orthogonal but complete basis of $\hat{\mathbf{k}}$, $\hat{\mathbf{k}}'$ and $\hat{\mathbf{g}} = \hat{\mathbf{k}} \times \hat{\mathbf{k}}'/|\hat{\mathbf{k}} \times \hat{\mathbf{k}}'|$. This results in,

$$\mathbf{T}^1_{\mathbf{kk}'} = \frac{(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}})(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}') - \hat{\mathbf{B}}\cdot\hat{\mathbf{k}}'}{(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')^2 - 1}\mathbf{T}^1_{\hat{\mathbf{B}}=\hat{\mathbf{k}}'} + \frac{(\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}')(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}') - \hat{\mathbf{B}}\cdot\hat{\mathbf{k}}}{(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')^2 - 1}\mathbf{T}^1_{\hat{\mathbf{B}}=\hat{\mathbf{k}}} + (\hat{\mathbf{B}}\cdot\hat{\mathbf{g}})\mathbf{T}^1_{\hat{\mathbf{B}}=\hat{\mathbf{g}}}. \tag{14}$$

The cases where $\hat{\mathbf{B}}$ is either along $\hat{\mathbf{k}}$ or $\hat{\mathbf{k}}'$ turn out to take the form,

$$T^1_{\sigma\sigma'}(\hat{\mathbf{B}} = \hat{\mathbf{k}}) = \frac{\pi}{\omega}\,[R_1(\cos\theta)\sigma + R_2(\cos\theta)\sigma'], \tag{15}$$

$$T^1_{\sigma\sigma'}(\hat{\mathbf{B}} = \hat{\mathbf{k}}') = \frac{\pi}{\omega}\,[R_1(\cos\theta)\sigma' + R_2(\cos\theta)\sigma] \tag{16}$$

with

$$R_1(\cos\theta) = -\frac{2W}{\pi}\sum_{J\geq 1}\frac{2J+1}{J(J+1)}\,[\mathscr{C}_J\pi_{J,1}(\cos\theta) + \mathscr{D}_J\tau_{J,1}(\cos\theta)], \tag{17}$$

$$R_2(\cos\theta) = -\frac{2W}{\pi}\sum_{J\geq 1}\frac{2J+1}{J(J+1)}\,[\mathscr{D}_J\pi_{J,1}(\cos\theta) + \mathscr{C}_J\tau_{J,1}(\cos\theta)]. \tag{18}$$

In Ref. [7] we gave an expression for $\mathbf{T}^1_{\sigma\sigma'}(\hat{\mathbf{B}}=\hat{\mathbf{g}})$ involving a double summation over the partial wave number J and the magnetic quantum number M. It is actually possible to do the summation over M explicitly, thus simplifying considerably the numerical evaluation. Indeed, if one expresses $\mathbf{T}^0$ with respect to a z-axis perpendicular to the scattering plane for a given partial wave J, one ends up with the following relation between the polynomials $\pi_{J,M}$ and $\tau_{J,M}$,

$$\pi_{J,1}(\cos\theta) = 2\sum_{J\geq M\geq 1}\left[\frac{(J-M)!}{(J+M)!}\,\tau_{J,M}(0)^2\cos(M\theta)\right] + \tau_{J,0}(0)^2, \tag{19}$$

$$\tau_{J,1}(\cos\theta) = 2\sum_{J\geq M\geq 1}\left[\frac{(J-M)!}{(J+M)!}\,\pi_{J,M}(0)^2\cos(M\theta)\right] + \pi_{J,0}(0)^2. \tag{20}$$

Upon performing the derivatives of these relations with respect to θ and comparing to the expression for $\mathbf{T}^1_{\sigma\sigma'}(\hat{\mathbf{B}}=\hat{\mathbf{g}})$ we find,

$$T^1_{\sigma\sigma'}(\hat{\mathbf{B}}=\hat{\mathbf{g}}) = \frac{\pi}{\omega}\,(Q_1(\theta)+\sigma\sigma' Q_2(\theta)) \tag{21}$$

with

$$Q_l(\theta) = -i\,\frac{\mathrm{d}}{\mathrm{d}\theta}\,R_l(\cos\theta) = i\sin\theta\,\frac{\mathrm{d}}{\mathrm{d}\cos\theta}\,R_l(\cos\theta), \quad l=1,2. \tag{22}$$

We are convinced that a rigorous group symmetry argument exists that relates the derivative of $T^1_{\sigma\sigma'}(\hat{\mathbf{B}}=\hat{\mathbf{k}})$ with respect to θ to $T^1_{\sigma\sigma'}(\hat{\mathbf{B}}=\hat{\mathbf{g}})$.

3.3. Particular case for $\boldsymbol{T}^1$ when $\hat{\boldsymbol{k}}=\hat{\boldsymbol{k}}'$ and $\hat{\boldsymbol{k}}=-\hat{\boldsymbol{k}}'$

The treatment in Section 3.2 becomes degenerate when $\hat{\mathbf{k}}$ and $\hat{\mathbf{k}}'$ are collinear, i.e. in forward or backward direction. In these cases, $\hat{\mathbf{B}}$ can still be expressed on a basis made of $\hat{\mathbf{k}}$ and of two vectors perpendicular to $\hat{\mathbf{k}}$. The contribution of these last two vectors has the same form as in $\mathbf{T}^1_{\sigma\sigma'}(\hat{\mathbf{B}}=\hat{\mathbf{g}})$ for $\theta=0$ or $\theta=\pi$, which vanishes. An alternative derivation consists to take the limit $\theta\to 0$ so that $R_1=R_2$ or $\theta\to\pi$ so that $R_1=-R_2$ in Eqs. (15)–(18). This yields,

$$R_1(1) = R_2(1) = -\frac{W}{\pi}\sum_{J\geq 1}(2J+1)(\mathscr{C}_J+\mathscr{D}_J), \tag{23}$$

and

$$R_1(-1) = -R_2(-1) = -\frac{W}{\pi}\sum_{J\geq 1}(-1)^{J+1}(2J+1)(\mathscr{C}_J-\mathscr{D}_J). \tag{24}$$

This means,

$$\mathbf{T}^1_{\mathbf{k},\mathbf{k}} = \boldsymbol{\Phi}\,\frac{2\pi}{\omega}\,R_1(1), \tag{25}$$

and

$$\mathbf{T}^1_{\mathbf{k},-\mathbf{k}} = \boldsymbol{\Phi}\,\frac{2\pi}{\omega}\,R_1(-1). \tag{26}$$

Both T-matrices contain the tensor $\mathbf{\Phi}$ introduced in Eq. (1) for the dielectric constant of the medium of the sphere. For these two cases, an operator can be associated with these T-matrices, which is $\mathbf{S_z}$ since we have chosen $\mathbf{B}$ along the z-axis. For $\mathbf{T}^1_{\mathbf{k},\mathbf{k}}$, the presence of the tensor $\mathbf{\Phi}$ is to be expected since we know that the forward scattering amplitude can be interpreted as an effective refractive index in a transmission experiment [9]. In the framework of an effective medium theory, the real part of Eq. (25) gives the Faraday effect whereas the imaginary part gives the magneto-dichroism (i.e. different absorption for different circular polarization) of an ensemble of Faraday-active scatterers.

4. Magneto-transverse scattering

From $\mathbf{T}^1$ matrix, we can compute how the magnetic field affects the differential scattering cross section (summed over polarization) as a function of the scattering angle. Its form can be guessed before doing any calculation at all, since it must satisfy mirror-symmetry and the reciprocity relation $\mathrm{d}\sigma/\mathrm{d}\Omega(\mathbf{k}\to\mathbf{k}',\mathbf{B}) = \mathrm{d}\sigma/\mathrm{d}\Omega(-\mathbf{k}'\to-\mathbf{k},-\mathbf{B})$. A magneto-cross-section proportional to $\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}$ or to $\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}'$ is parity forbidden since $\mathbf{B}$ is a pseudo-vector. Together with the rotational symmetry of the sphere the only possibility is

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\mathbf{k}\to\mathbf{k}',\mathbf{B}) = F_0(\cos\theta) + \det(\hat{\mathbf{B}},\hat{\mathbf{k}},\hat{\mathbf{k}}')F_1(\cos\theta), \tag{27}$$

where $\det(\mathbf{A},\mathbf{B},\mathbf{C}) = \mathbf{A}\cdot(\mathbf{B}\times\mathbf{C})$ is the scalar determinant constructed from these three vectors. The second term in Eq. (27) will be called the magneto-cross section.

The magneto-cross section implies that there may be more photons scattered “upwards” than “downwards”, both directions being defined with respect to the magneto-transverse vector $\hat{\mathbf{k}}\times\hat{\mathbf{B}}$ perpendicular to both incident wave vector and magnetic field. An easy calculation yields,

$$\Delta\sigma = \sigma_{\mathrm{up}} - \sigma_{\mathrm{down}} = \pi\int_0^\pi \mathrm{d}\theta\,\sin^3\theta F_1(\cos\theta). \tag{28}$$

A non-zero value for $\Delta\sigma$ will be referred to as a *Photon Hall Effect* (PHE).

For Rayleigh scatterers, the above theory simplifies dramatically because one only needs to consider the first partial wave of $J = 1$ and the first terms in a development in powers of y (since $y \ll 1$). From Eqs. (6) and (7) we find that, $\mathscr{C}_1 = -2y^3/m^2(2+m^2)^2$ and $\mathscr{D}_1 = -y^5/45m^4$. We can keep only $\mathscr{C}_1$ and drop $\mathscr{D}_1$ as a first approximation. Adding all the contributions of Eqs. (14) and (11), we find, in the linear base

$$\mathbf{T}_{\mathbf{k},\mathbf{k}'} = \begin{pmatrix} t_0\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}' + it_1\hat{\mathbf{B}}\cdot(\hat{\mathbf{k}}\times\hat{\mathbf{k}}') & -it_1\hat{\mathbf{B}}\cdot\hat{\mathbf{k}} \\ it_1\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}' & t_0 \end{pmatrix}. \tag{29}$$

where $t_0 = -6\mathrm{i}\pi a_1^\dagger/\omega$ and $t_1 = -6C_1W/\omega$. This form agrees with the Rayleigh point-like scatterer discussed in Ref. [12].

A magnetic field breaks the rotational symmetry of the particle. If it is contained in the scattering plane, Eq. (29) shows that we must have a non-zero value for S_3 and S_4 as opposed to the case when no magnetic field is applied. This property still holds for a Mie scatterer, the difference being only

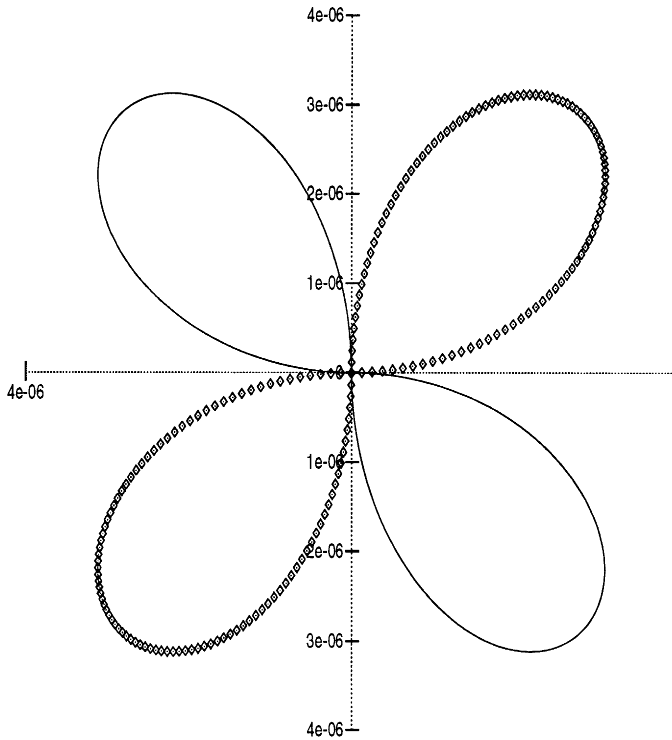

Fig. 2. Magneto-scattering cross section $F_1(\theta)$ for a Rayleigh scatterer. The solid line is a positive correction and the points denote a negative correction. The curve has been normalized by the parameter W. No *Hall effect* is expected in this case because the projection onto the y-axis of both corrections cancel.

present in the angular dependence of the elements of the amplitude matrix. A magnetic field also violates the standard reciprocity principle as can be seen on Eq. (9). This implies that $S_3 + S_4$ is non zero for the backward direction $\theta = \pi$. The relation $S_3 + S_4 = 0$ for the backward direction was derived by Van Hulst, but does not apply when a magnetic field is present. In fact, the magnetic field imposes that $S_3 = S_4$ at backscattering. This is readily confirmed by the Rayleigh particle, for which Eq. (29) implies that $S_3 + S_4 = 2S_3 = -2it_1\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}$ for $\theta = \pi$.

Eq. (29) yields $F_1(\cos\theta) \sim VB\cos\theta/k$ so that Eq. (28) gives $\Delta\sigma = 0$. The magneto scattering cross section is shown in Fig. 2 for a Rayleigh scatterer and in Fig. 3 for a Mie scatterer for which a non zero value of $\Delta\sigma$ is seen to survive.

5. Stokes parameters

To describe the flux and polarization, a four-dimensional Stokes vector (I, Q, U, V) can be introduced [9]. The general relation between scattered Stokes vector and incoming Stokes vector is

$$(I, Q, U, V)_{\text{out}} = \mathbf{F}(I, Q, U, V)_{\text{inc}}. \tag{30}$$

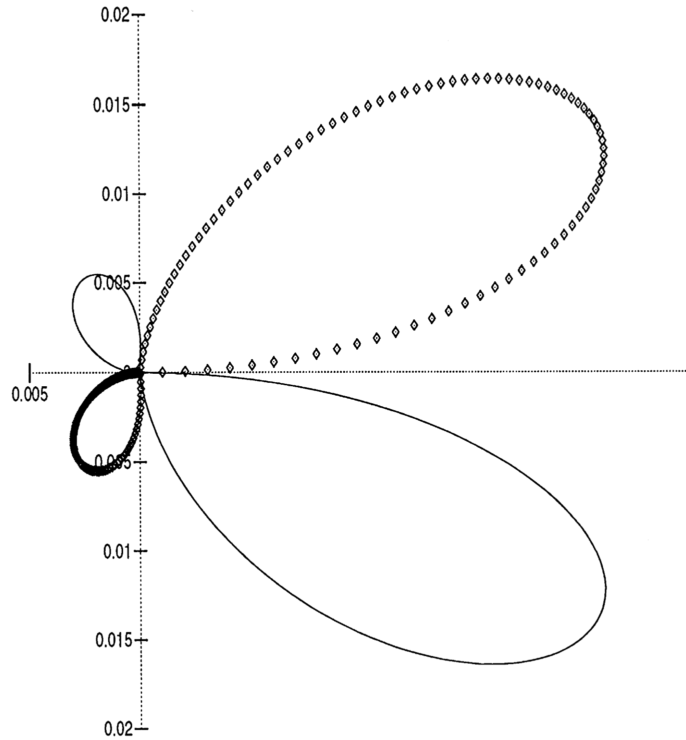

Fig. 3. Magneto-scattering cross section $F_1(\theta)$ for a Mie scatterer of size parameter $x = 2$. The curve has been normalized by the parameter W. The solid line is for positive correction and the points are the negative correction. Now, there is a *Hall effect*: $\Delta\sigma \neq 0$.

For a sphere and without a magnetic field, the F-matrix is well known and equals [9],

$$F^0_{ij} = \frac{1}{k^2r^2}\begin{pmatrix} F_{11} & F_{12} & 0 & 0 \\ F_{12} & F_{11} & 0 & 0 \\ 0 & 0 & F_{33} & F_{34} \\ 0 & 0 & -F_{34} & F_{33} \end{pmatrix}, \tag{31}$$

where

$$\begin{aligned} F_{11} &= (|S_1|^2 + |S_2|^2)/2, \\ F_{12} &= (-|S_1|^2 + |S_2|^2)/2, \\ F_{33} &= (S_2^*S_1 + S_2S_1^*)/2, \\ F_{34} &= i(-S_2^*S_1 + S_2S_1^*)/2. \end{aligned} \tag{32}$$

Among these four parameters only three are independent since $F_{11}^2 = F_{12}^2 + F_{33}^2 + F_{34}^2$. The presence of the many zeros in Eq. (31) is a consequence of the fact that the amplitude matrix in Eq. (10) is diagonal for one Mie scatterer. It is in fact much more general. The form of Eq. (31) still holds for an ensemble of randomly oriented particles with an internal plane of symmetry (such as spheroids for instance) [13]. In that case, the averaging is essential to get the many zeros in Eq. (31). It also holds for a single anisotropic particle in the Rayleigh–Gans approximation [14,15].

For the Mie case, the anisotropy has two consequences: the F-elements that were zero for an isotropic particle may take finite values, and they may depend on the azimuthal angle ϕ. When a magnetic field is applied perpendicular to the scattering plane, corrections will appear in the diagonal terms of the amplitude matrix. We will use the vector H to denote them. When a magnetic field is applied in the scattering plane, the amplitude matrix becomes off-diagonal, which will fill up the zeros in F^0. We will use the vector G to denote these new terms.

If we call F^1 the first-order magnetic correction to the F-matrix one finds,

$$F^1_{ij} = \frac{1}{k^2 r^2} \begin{pmatrix} H_{11} & H_{12} & \Re e G_3 & -\Im m G_3 \\ H_{12} & H_{11} & \Re e G_4 & -\Im m G_4 \\ \Re e G_1 & \Re e G_2 & H_{33} & H_{34} \\ \Im m G_1 & \Im m G_2 & -H_{34} & H_{33} \end{pmatrix}. \tag{33}$$

When $\hat{\mathbf{B}}$ is directed along $\hat{\mathbf{k}}$, the G terms are given by

$$G_{\hat{\mathbf{B}}=\hat{\mathbf{k}}} \begin{cases} G_1 = (S_1^* R_1^* - S_2 R_2)/2 \\ G_2 = (-S_1^* R_1^* - S_2 R_2)/2 \\ G_3 = (-S_1^* R_2^* + S_2 R_1)/2 \\ G_4 = (S_1^* R_2^* + S_2 R_1)/2. \end{cases} \tag{34}$$

The general case (forward and backward directions excluded) has

$$\mathbf{G} = \frac{(\hat{\mathbf{B}}\cdot\hat{\mathbf{k}})(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}') - \hat{\mathbf{B}}\cdot\hat{\mathbf{k}}'}{(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')^2 - 1}\,\mathbf{G}_{\hat{\mathbf{B}}=\hat{\mathbf{k}}'} + \frac{(\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}')(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}') - \hat{\mathbf{B}}\cdot\hat{\mathbf{k}}}{(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')^2 - 1}\mathbf{G}_{\hat{\mathbf{B}}=\hat{\mathbf{k}}}, \tag{35}$$

and $G_{\hat{\mathbf{B}}=\hat{\mathbf{k}}'}$ is obtained from $G_{\hat{\mathbf{B}}=\hat{\mathbf{k}}}$ by exchanging R_1 and R_2 in Eq. (34) like in Eqs. (15) and (16). Finally, we need,

$$\mathbf{H} = (\hat{\mathbf{B}}\cdot\hat{\mathbf{g}})\mathbf{H}_{\hat{\mathbf{B}}=\hat{\mathbf{g}}} \tag{36}$$

with

$$\mathbf{H}_{\hat{\mathbf{B}}=\hat{\mathbf{g}}} \begin{cases} H_{11} = -\Im m(S_1 Q_1 + S_2 Q_2)/2 \\ H_{12} = -\Im m(-S_1 Q_1 + S_2 Q_2)/2 \\ H_{33} = \Im m(-S_1 Q_2 - S_2 Q_1)/2 \\ H_{34} = \Re e(S_1 Q_2 - S_2 Q_1)/2. \end{cases} \tag{37}$$

The F-matrix defined in Eq. (30) can contain at most 7 independent constants, resulting from the 8 constants in the amplitude matrix minus an irrelevant phase. Our F^1-matrix has 12 coefficients (4 for the H vector and 8 for the G vector). Therefore, 5 relations must exist between these 12 coefficients. These relations have not been explicitly derived.

We can write all the expressions above in a very compact way using the basis of the Pauli matrices [16]

$$F^0_{ij} = \tfrac{1}{2}Tr(A^{0\dagger}\sigma_i A^0 \sigma_j),$$
$$F^1_{ij} = \tfrac{1}{2}Tr(A^{1\dagger}\sigma_i A^0 \sigma_j) + \tfrac{1}{2}Tr(A^{0\dagger}\sigma_i A^1 \sigma_j), \tag{38}$$

where Tr is the trace of the matrix, the superscript $\dagger$ denotes Hermite conjugation, σ_i are Pauli matrices and A^0, A^1 are zeroth and first-order correction in the amplitude matrix defined as the T-matrix from Eq. (10).

If the incident light is unpolarized, the Stokes vector for the scattered light is simply equal to the first column of the F-matrix in Eq. (33). For instance when $\hat{\mathbf{B}}$ is directed along $\hat{\mathbf{k}}$, the magnetic field will only affect $U = F^1_{31}$ and the circular polarization $V = F^1_{41}$ which would be zero when no magnetic field is applied. We choose to normalize the matrix elements F^1_{ij} that quantify the deviation of the polarization from the isotropic case by the flux F^0_{11} without magnetic field. In Fig. 4 we plotted these normalized matrix elements for the cases where $\hat{\mathbf{B}}$ is directed along $\hat{\mathbf{k}}$ and where $\hat{\mathbf{B}}$ is directed along $\hat{\mathbf{g}}$. We observe that off-diagonal F-elements such as F^1_{12} and F^1_{41}, are generally more important in the angle region of 140–170°, and increase with the size parameter. In this region, these Stokes parameters seem to be very sensitive to anisotropy as also found from studies of Stokes parameters of quartz particles [16].

The F-matrix of spherical scatterers in Eq. (31) contains 8 zeros among its 16 elements. This property persists for an ensemble of randomly oriented non-spherical particles having a plane of symmetry because of the averaging over all the orientations. In a magnetic field even spherical scatterers can have a non-zero value for these 8 elements. Furthermore, we have good reasons to believe that our theory made for spheres in a magnetic theory should also apply to an ensemble of randomly oriented non-spherical particles in a magnetic field, since the magnetic field direction is the same for all the particles.

We have chosen the size distribution [17] and optical parameters of a reported experiment [18], when no magnetic field is present, but in which all the matrix elements of F^0 were measured and found to be in good agreement with the theoretical evaluation from Eq. (31). For water, the parameter $W \approx 2.4 \times 10^{-6}$ for a magnetic field of 1 T. From Fig. 4, we can therefore expect a modification of the order of 2.4×10^{-6} in the region near backward scattering for $F^1_{31}\,(B)/F^1_{11}(B=0)$ when $\hat{\mathbf{B}}$ is directed along $\hat{\mathbf{k}}$. The magneto-optical effects on polarization are very small. Nevertheless, they may become significant in multiple scattering, which usually tends to depolarize completely the light.

5.1. Forward and backward directions

When no magnetic field is present, the situations for $\theta = 0$ or $\theta = \pi$ are similar because the scattering plane is undefined in both cases. We also have $H = 0$ by Eq. (36). The remaining contribution is therefore only determined by the G-vector, and the final result reads for $\theta = 0$,

$$F_{\theta=0} = \frac{\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}}{k^2r^2}\begin{pmatrix} 0 & 0 & 0 & -\Im m(z) \\ 0 & 0 & \Re e(z) & 0 \\ 0 & -\Re e(z) & 0 & 0 \\ -\Im m(z) & 0 & 0 & 0 \end{pmatrix} \tag{39}$$

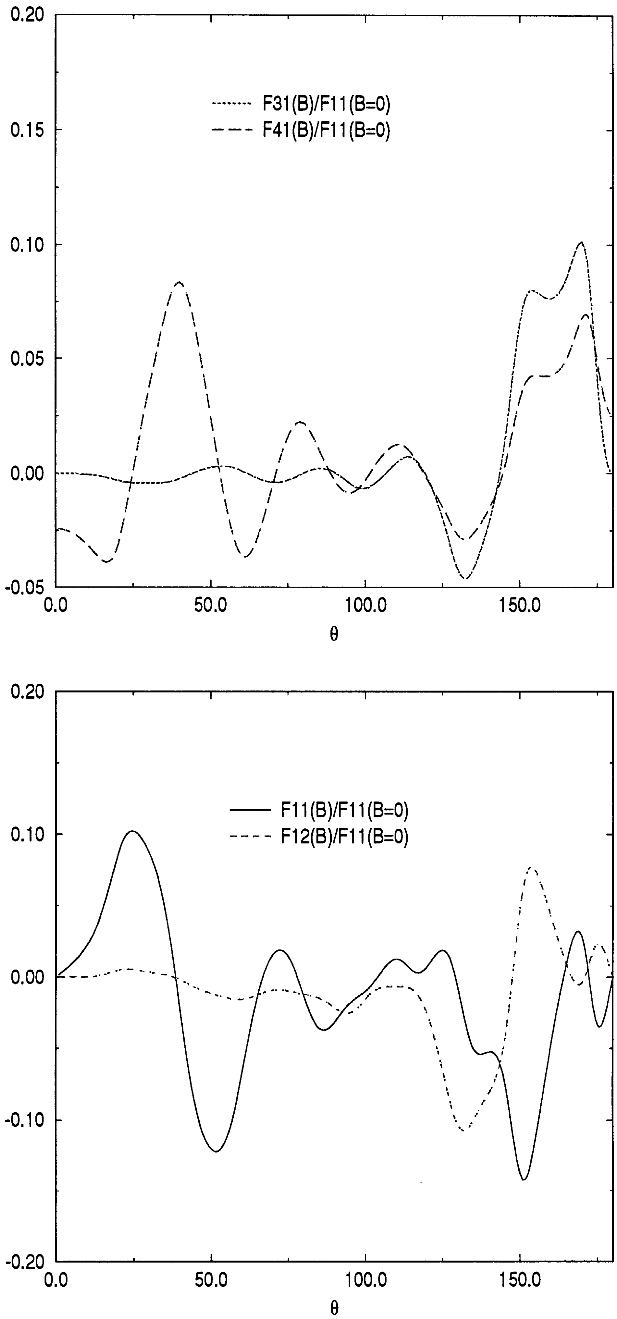

Fig. 4. Scattering matrix elements $F^1_{31}(B)/F^1_{11}(B=0)$ and $F^1_{41}(B)/F^1_{11}(B=0)$ of an ensemble of water droplets as a function of scattering angle for $\hat{\mathbf{B}}$ directed along $\hat{\mathbf{k}}$ (top), and $F^1_{11}(B)/F^1_{11}(B=0)$ and $F^1_{12}(B)/F^1_{11}(B=0)$ for $\hat{\mathbf{B}}$ directed along $\hat{\mathbf{g}}$ (bottom). The refractive index is 1.332, a lognormal size distribution has been used with $r_{\mathrm{eff}} = 0.75$ μm and $\sigma_{\mathrm{eff}} = 0.45$ and $\lambda = 632.8$ nm. The curve has been displayed for $W = 0.1$.

with $z = S_1(1)R_1(1)$. For $\theta = \pi$,

$$F_{\theta=\pi} = \frac{\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}}{k^2r^2}\begin{pmatrix} 0 & 0 & 0 & \Im m(z') \\ 0 & 0 & -\Re e(z') & 0 \\ 0 & -\Re e(z') & 0 & 0 \\ -\Im m(z') & 0 & 0 & 0 \end{pmatrix} \tag{40}$$

with $z' = S_1(-1)R_1(-1)$.

The functions $R_1(1)$ and $R_1(-1)$ defined in Eqs. (17) and (18) are very similar to $S_1(1)$ and $S_1(-1)$. Both F-matrices contain only two real-valued independent parameters as do the corresponding T matrices. For unpolarized incident light only the Stokes parameter $V = F^1_{41}(B)$ of these matrices is non-zero. In Fig. 4, all the curves are zero for $\theta = 0$ and $\theta = \pi$ except the one of $F^1_{41}(B)/F^1_{11}(B=0)$. In other words, unpolarized incident light will produce partially circularly polarized light (the degree of circular polarization being precisely $F^1_{41}(B)/F^0_{11}(B=0)$) for $\hat{\mathbf{B}}$ directed along $\hat{\mathbf{k}}$ in the forward and backward directions. This can be understood from the fact that the effective index that one can define from Eq. (25) suffers from magneto-dichroism (i.e. different absorption for different circular polarization).

The modified reciprocity relation in the presence of a magnetic field was expressed for the amplitude matrix in Eq. (9). For the F-matrix it implies exactly the different signs in the matrix elements of Eq. (40) with respect to Eq. (39).

6. Summary and outlook

We have shown that the theory developed for magneto-active Mie scatterers so far is consistent with the former results concerning predictions of the light scattering by Rayleigh scatterers in a magnetic field. Our perturbative theory provides quantitative predictions concerning the *Photonic Hall Effect* for one single Mie sphere, such as the scattering cross section, the dependence on the size parameter or on the index of refraction.

Using the magneto-correction to the T-matrix we have derived the Stokes parameters for the light scattered from a single sphere in a magnetic field. We have distinguished two main cases. Either the magnetic field is perpendicular to the scattering plane and there will be corrections to the usual non-zero Stokes parameters, or when the magnetic field is in the scattering plane, the corrections fill up the F-matrix elements which were previously zero. We have discussed the particular cases of forward and backward scattering.

We hope that these results will be useful in comparing them to the situation in multiple scattering. Even after many scattering events, we suspect that the presence of a magnetic field prevents the Stokes parameters U, V and Q to be zero. In single scattering, their order of magnitude is controlled by the parameter $W = V_0 B\lambda$. In multiple scattering, however, this parameter must be replaced by fV_0Bl^*, where f is the volume fraction of the scatterers and l^* the transport mean free path, and $fl^* \gg 1$. We expect to find more significant effects in this case.

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Appendix A. Derivation of reciprocity and parity relations

In the indices for polarization in the T-matrix, the state of helicity σ is to be referred to the direction of the wave vector immediately close to it. In Eqs. (8) and (9), $T_{-\mathbf{k}\sigma,-\mathbf{k}'\sigma'}$ for instance really means $T_{-\mathbf{k}\sigma(-\mathbf{k}),-\mathbf{k}'\sigma'(-\mathbf{k}')}$. To derive these equations, we start from Eq. (5) in which we change both incoming and outcoming wave vectors into their opposite:

$$\mathbf{T}^1_{-\mathbf{k}\sigma,-\mathbf{k}'\sigma'} = \sum_{J,M,\lambda} (-M)[\alpha_{J,\lambda}\mathbf{Y}^{\lambda}_{J,M}(-\hat{\mathbf{k}})\cdot\chi_{\sigma(-\mathbf{k})}(-\mathbf{k})\mathbf{Y}^{\lambda *}_{J,M}(-\hat{\mathbf{k}}')\cdot\chi_{\sigma'(-\mathbf{k}')}(-\mathbf{k}')], \tag{41}$$

where $\alpha_{J,\lambda}$ is a well-known coefficient, $\chi_{\sigma(\mathbf{k})}$ is the eigenfunction of the operator $\mathbf{S}\cdot\mathbf{k}$ with eigenvalue $\sigma(\mathbf{k})$ the helicity. $\mathbf{S}$ is a spin one operator acting on three-dimensional vectors. The summation is to be performed for $\lambda = e, m$ only, which are associated with the two transverse components of the given vector spherical harmonics. $\mathbf{Y}^{\lambda}_{JM}(\mathbf{k})$ is a well-defined linear combination of $\mathbf{Y}^{M}_{J,J}(\mathbf{k})$, $\mathbf{Y}^{M}_{J,J-1}(\mathbf{k})$ and $\mathbf{Y}^{M}_{J,J+1}(\mathbf{k})$ that obeys [10]

$$\mathbf{Y}^{\lambda}_{JM}(\mathbf{k})\cdot\mathbf{k} = 0, \qquad \lambda = e,m.$$

We now use the relations,

$$\begin{aligned}\mathbf{Y}^{e}_{J,M}(-\hat{\mathbf{k}}) &= (-1)^{J+1}\mathbf{Y}^{e}_{J,M}(\hat{\mathbf{k}}),\\ \mathbf{Y}^{m}_{J,M}(-\hat{\mathbf{k}}) &= (-1)^{J}\mathbf{Y}^{m}_{J,M}(\hat{\mathbf{k}}).\end{aligned} \tag{42}$$

The eigenfunctions $\chi_{\sigma(\mathbf{k})}$ also change under parity since

$$\chi_{\sigma(-\mathbf{k})}(-\mathbf{k}) = -\chi_{\sigma(\mathbf{k})}(\mathbf{k}).$$

Because of this additional minus sign, the parities of the vector spherical harmonics are in fact,

$$\begin{aligned}\mathbf{P}\mathbf{Y}^{e}_{J,M} &= (-1)^{J}\mathbf{Y}^{e}_{J,M},\\ \mathbf{P}\mathbf{Y}^{m}_{J,M} &= (-1)^{J+1}\mathbf{Y}^{m}_{J,M}.\end{aligned} \tag{43}$$

The parity symmetry relation of Eq. (8) follows from the application of these relations into Eq. (41). The proof of the reciprocity symmetry relation of Eq. (9) is similar, where now the following relations are necessary:

$$\mathbf{Y}^{\lambda *}_{J,M} = (-1)^{J+M}\mathbf{Y}^{\lambda}_{J,-M} \tag{44}$$

for $\lambda = e, o, m$ and

$$\chi^{*}_{\sigma(\mathbf{k})}(\mathbf{k}) = \chi_{-\sigma(\mathbf{k})}(\mathbf{k}).$$

The change of sign of $\mathbf{B}$ is provided by the M factor in Eq. (41) as surmised.

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