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## THE PHOTON'S MAGNETIC FIELD $\mathbf{B}_\Pi$ : THE MAGNETIC NATURE OF ANTISYMMETRIC LIGHT SCATTERING

### I. INTRODUCTION

The classical intensity of electromagnetic radiation is a tensor ( $I_{ij}$ ) proportional to the tensor product  $E_i E_j^*$  (Refs. 1-3). Here  $E_i$  is a component of the electric field strength and  $E_i^*$  denotes its complex conjugate.<sup>4</sup> In free space, the scalar part of the intensity is

$$I_0 = \varepsilon_0 c E_0^2 \quad (1)$$

where  $\varepsilon_0$  is the free space permittivity,  $c$  the speed of light and  $E_0$  the scalar amplitude of the electric field strength of the electromagnetic plane wave.<sup>5,6</sup> The vector part of  $E_i E_j^*$  is conveniently expressed as the conjugate vector cross product:

$$\mathbf{\Pi}^{(\Lambda)} = \mathbf{E} \times \mathbf{E}^* = 2E_0^2 \mathbf{i k} \quad (2)$$

which is purely imaginary as a consequence of the fact that  $E_i E_j^*$  is a Hermitian tensor of rank two.<sup>7,8</sup> It has recently been shown<sup>9-12</sup> that the conjugate product  $\mathbf{\Pi}^{(\Lambda)}$  is directly proportional to a novel magnetostatic flux density vector  $\mathbf{B}_\Pi$  of the classical electromagnetic plane wave:

$$\mathbf{B}_\Pi = \frac{\mathbf{\Pi}^{(\Lambda)}}{2E_0 c i} = B_0 \mathbf{k} = \left( \frac{I_0}{\varepsilon_0 c^3} \right)^{1/2} \mathbf{k} = \left( \frac{|\mathbf{N}|}{2\varepsilon_0 c^3} \right)^{1/2} \mathbf{k} \quad (3)$$

Here  $\mathbf{k}$  is a unit axial vector in the propagation axis of the plane wave,  $B_0$  is the plane wave's scalar magnetic flux density amplitude, and  $\mathbf{N}$  is the Poynting vector:

$$\mathbf{N} = \frac{1}{\mu_0} \mathbf{E} \times \mathbf{B}^* \quad (4)$$

Remarkably, the vector  $\mathbf{B}_\Pi$ , a flux of magnetic density (in tesla) independent of the phase of the plane wave, has been overlooked in the long and illustrious history of the classical theory of fields,<sup>8</sup> whereas its close relative  $\mathbf{N}$ , a flux of energy density, has been well known for many years. Furthermore, the interpretation of  $\mathbf{B}_\Pi$  in the quantum theory of fields leads straightforwardly<sup>10</sup> to the conclusion that the photon generates on the most fundamental level a magnetic flux density operator

$$\hat{B}_\Pi = B_0 \frac{\int}{\hbar} \quad (5)$$

directly proportional to its angular momentum  $\hat{J}$ , a well-known boson operator.<sup>13</sup> Here  $\hbar$  is the unit of angular momentum in quantum mechanics, the reduced Planck constant.

Quite generally, therefore, the ubiquitous antisymmetric part of the electromagnetic intensity (denoted by  $I_{ij}$ ) can be rewritten in terms of the novel magnetic vector  $\mathbf{B}_\Pi$ , leading immediately to novel insights about all processes in physical optics that depend on  $I_{ij}$ .

In this paper we illustrate this conclusion with reference to the antisymmetric part of Rayleigh scattering from molecular liquids, a process first considered by Placzek<sup>14</sup> in 1934. Section II defines the antisymmetric part of the scattered intensity in terms of the scattered  $\mathbf{B}_\Pi$  vector, adapting the arguments of Knast and Kielich.<sup>7</sup> Section III continues the development in terms of Rayleigh refringent scattering theory, used to relate the incoming and scattered magnetic vectors  $\mathbf{B}_\Pi$ . Section IV is a discussion of these results, leading to the conclusions that antisymmetric light scattering in general can be reinterpreted fundamentally as a purely magnetic process, whereby the incoming  $\mathbf{B}_\Pi$  (or  $\hat{B}_\Pi$  of quantum field theory) interacts with the molecular ensemble forming the scattering volume, and is scattered as the vector  $\mathbf{B}_{\Pi S}$ . The two magnetic vectors  $\mathbf{B}_\Pi$  and  $\mathbf{B}_{\Pi S}$  are related through the molecular property tensors of the scattering volume.

## II. DEFINITION OF THE SCATTERED $\mathbf{B}_{\Pi S}$ VECTOR

It is convenient to define the scattered  $\mathbf{B}_{\Pi S}$  vector in terms of the development of Knast and Kielich,<sup>7</sup> who have also tabulated extensively the magnetic point group symmetries of the antisymmetric part of the molecular and atomic polarizability. In so doing we define the intensity tensor of the incoming light as

$$I_{ij} = \epsilon_0 c E_i E_j^* \quad (6)$$

from which it follows that the vector part of the intensity, the antisymmetric component  $I_{ij}^-$  of Knast and Kielich<sup>7</sup> can be expressed simply as the purely imaginary axial vector:

$$\begin{aligned} \mathbf{I}^- &\equiv \frac{1}{2} \epsilon_{ijk} (I_{ij} - I_{ji}) = i I_0 \mathbf{k} \\ &= \epsilon_0 c^2 E_0 i \mathbf{B}_{\Pi} \end{aligned} \quad (7)$$

where  $\mathbf{k}$  is a unit axial vector in the propagation direction. From these definitions we deduce immediately that the incoming  $\mathbf{B}_{\Pi}$  vector is proportional to the square root of the incoming  $I_0$ :

$$\mathbf{B}_{\Pi} = \left( \frac{I_0}{\epsilon_0 c^2 E_0} \right) \mathbf{k} = \left( \frac{I_0}{\epsilon_0 c^3} \right)^{1/2} \mathbf{k} \quad (8)$$

The antisymmetric part of the scattered light intensity tensor is defined by Knast and Kielich to be:

$$I_{ijS}^-(t) = \left( \frac{\omega_0}{c} \right)^4 \langle M_i(\mathbf{r}, t_0) M_j^*(\mathbf{r}, t) \rangle_{t_0}^- \quad (9)$$

where

$$M_i(\mathbf{r}, t_0) = \mu_i^{(p)}(t_0) \exp[i(\Delta \mathbf{k} \cdot \mathbf{r}(t_0))] \quad (10)$$

where  $\mu_i^{(p)}$  is the  $i$ th component of the electric dipole moment induced in a molecule  $p$  by the light's electric field strength vector, as usual in the theory of scattering.<sup>15,16</sup> The quantity  $\Delta \mathbf{k}$  is a difference in wave vectors of incident and scattered light, as usual, and the summation extends over all the  $N$  molecules of the volume. The angular brackets in Eq. (9) denote a time-correlation function<sup>17</sup> of the fluctuating quantity  $M_i$ . It is well known

from the theory of nonequilibrium statistical mechanics<sup>17</sup> that the normalized time-correlation function starts at unity and evolves to zero with time  $t$ . Its Fourier transform is a spectral function of angular frequency  $\omega$ . In Rayleigh scattering<sup>15-17</sup> the spectrum is the experimental Rayleigh band-shape, which is Fourier transformed to give the time-correlation function. The scattered antisymmetric intensity  $I_{ijS}^-(k)$  is therefore frequency dependent, and forms the antisymmetric part of the Rayleigh scattering spectrum. However,  $I_{ijS}^-(t)$  is directly proportional to the scattered  $\mathbf{B}_{\Pi S}$  vector, (or in quantum-field theory<sup>10</sup> the scattered flux density operator  $\hat{B}_{\Pi S}$ ) and we reach the significant conclusion that the spectrum of antisymmetric Rayleigh scattering is a graph of the scattered  $\beta_{\Pi S}(\omega)$  plotted against the change in frequency  $(\omega - \omega_0)$ , where  $\omega_0$  is the incoming laser frequency. Antisymmetric Rayleigh scattering is therefore a process that can be described entirely in terms of the vector  $\mathbf{B}_{\Pi}$ .

This conclusion can be underlined by expressing the fundamental equation (9) in terms of a mean magnetic dipole moment  $\langle \mathbf{m}_S(t) \rangle$ , formed at time  $t$  from the antisymmetric part of the time correlation function  $\langle M_i(\mathbf{r}, t_0) M_j^*(\mathbf{r}, t) \rangle_{t_0}$ . This development is based on the relation

$$\langle \mathbf{m}_S(t) \rangle = \xi \langle \mathbf{M}(\mathbf{r}, t_0) \times \mathbf{M}^*(\mathbf{r}, t) \rangle_{t_0} \quad (11)$$

where  $\xi$  is a proportionality coefficient; i.e., the vector cross product of two nonidentical electric dipole moment vectors is proportional to a magnetic dipole moment. This conclusion can be illustrated by the following simple model.

Express the electric dipole moments  $\boldsymbol{\mu}$  and  $\boldsymbol{\mu}^*$  as products of electronic charge  $e$  and position vectors  $\mathbf{r}$  and  $\mathbf{r}^*$ . Then the cross product can be expressed as an area

$$\boldsymbol{\mu} \times \boldsymbol{\mu}^* = e^2 \mathbf{r} \times \mathbf{r}^* = e^2 A \mathbf{k} \quad (12)$$

Considering the simple model of the motion of charge with instantaneous linear velocity  $v$  around a circle of radius  $r$ , the magnetic dipole moment is known<sup>18</sup> to be proportional to the product  $IA$ , where  $A$  is the area of the circle, and  $I$  is the quantity  $ev/(2\pi r)$ . It follows that, in general, a magnetic dipole moment is proportional to area, and therefore to the cross product of  $\boldsymbol{\mu}$  and  $\boldsymbol{\mu}^*$ . The same conclusion is derived for the cross product of transition electric dipole moments by Atkins and Miller.<sup>19</sup> It is also a consequence of the fact that antisymmetric electric polarizability is proportional to the cross product of transition electric dipole moments, and therefore has the same symmetry<sup>20</sup> as a magnetic dipole moment.

From these considerations, we reach the

$$\mathbf{m}_S(t) = \frac{1}{\xi} \left( \frac{c}{\omega_0} \right)^4 \varepsilon_0 c^2 E_{0s} \mathbf{B}_{\Pi S}(t) \quad (13)$$

showing that the magnetic dipole moment  $\mathbf{m}_S(t)$  is proportional to the scattered  $\mathbf{B}_{\Pi S}(t)$  vector at time  $t$ . Fourier transformation leads immediately to the conclusion that the magnetic dipole moment at frequency  $\omega$  is proportional to the scattered  $\mathbf{B}_{\Pi S}(\omega)$  vector at the frequency  $\omega$  of the antisymmetric Rayleigh scattering spectrum.

### III. REFRACTIVE SCATTERING APPROACH

In this section we adapt straightforwardly Rayleigh refringent scattering theory to provide an expression linking the incoming  $\mathbf{B}_{\Pi}$  and the scattering  $\mathbf{B}_{\Pi S}$  in terms of a parameter  $\Xi_Z$ , which is defined in terms of the molecular property tensors of the scattering volume. The scattered light intensity tensor is defined in semiclassical Rayleigh refringent scattering theory by<sup>20</sup>

$$\begin{aligned} I_{\alpha\beta}^{(S)} &= \varepsilon_0 c E_{\alpha}^{(S)} E_{\beta}^{*(S)} \\ &= \varepsilon_0 c \left( \frac{\omega^2 \mu_0}{4\pi R} \right)^2 a_{\alpha\gamma} a_{\beta\delta}^* E_{\gamma}^{(0)} E_{\delta}^{*(0)} \end{aligned} \quad (14)$$

where

$$E_{\alpha}^{(S)} = \frac{\omega^2 \mu_0}{4\pi R} \exp \left[ i\omega \left( \frac{R}{c} - t \right) \right] a_{\alpha\beta} E_{\beta}^{(0)} \quad (15)$$

is the scattered electric field detected in the wave zone at a point  $d$  at distance  $R$  from the molecular origin. The origin of scattered light is considered to be the characteristic radiation field generated by the oscillating electric and magnetic multipole moments induced in a molecule by the electromagnetic fields of the incident light wave. Here  $a_{\alpha\beta}$  is the scattering tensor, a molecular property of the scattering volume for particular incident and scattered directions given by unit vectors  $\mathbf{n}^{(0)}$  and  $\mathbf{n}^{(S)}$ . In Eq. (14)  $\omega$  is the angular frequency of the incoming wave, whose electric field strength vector is denoted  $\mathbf{E}^{(0)}$ , so that the scattered intensity tensor  $I_{\alpha\beta}^{(S)}$  is

1 terms of the incident intensity tensor  $I_{\gamma\delta}^{(0)}$  by

$$I_{\alpha\beta}^{(S)} = \left( \frac{\omega^2 \mu_0}{4\pi R} \right)^2 a_{\alpha\gamma} a_{\beta\delta}^* I_{\gamma\delta}^{(0)} \quad (16)$$

It is immediately clear, therefore, that the incident  $\mathbf{B}_{\Pi}$  in antisymmetric scattering can be expressed in terms of the scattered  $\mathbf{B}_{\Pi S}$  in a similar way. Thus, we arrive at the conclusion that antisymmetric light scattering, in general, is a process whereby the incoming  $\mathbf{B}_{\Pi}$  is transformed into a scattered  $\mathbf{B}_{\Pi S}$ ; i.e., antisymmetric Rayleigh scattering is a purely magneto-optic process. This argument is developed by consideration of the antisymmetric (vector) part of the scattered intensity tensor

$$\begin{aligned} I_{\varepsilon}^{(AS)} &= \varepsilon_0 c \Pi_{\varepsilon}^{(AS)} \\ \Pi_{\varepsilon}^{(AS)} &= \frac{1}{2} \left( \frac{\omega^2 \mu_0}{4\pi R} \right)^2 \varepsilon_{\alpha\beta\varepsilon} \left( E_{\alpha}^{(S)} E_{\beta}^{*(S)} - E_{\beta}^{(S)} E_{\alpha}^{*(S)} \right) \end{aligned} \quad (17)$$

For the  $Z$  component

$$\Pi_Z^{(AS)} = 2 E_0^{(0)2} i \Xi_Z \quad (18)$$

where

$$\begin{aligned} \Xi_Z &= \frac{\omega^4 \mu_0^2}{32\pi^2 R^2 i} \left[ (a_{XX} a_{YY}^* - a_{YX} a_{XX}^*) + (a_{XY} a_{YY}^* - a_{YY} a_{XY}^*) \right. \\ &\quad \left. + i(a_{XX} a_{YY}^* - a_{YX} a_{XX}^*) - i(a_{XY} a_{YX}^* - a_{YY} a_{XX}^*) \right] \end{aligned} \quad (19)$$

is in general a complex quantity. Using the result

$$\Pi_Z^{(A)} = 2 E_0^{(0)2} i \quad (20)$$

it follows that

$$\Pi_Z^{(AS)} = \Pi_Z^{(A)} \Xi_Z \quad (21)$$

and that

$$\mathbf{B}_{\Pi SZ} = \Xi_Z \mathbf{B}_{\Pi Z} \quad (22)$$

For forward scattering, there is no component of  $\mathbf{B}_{\Pi S}$  other than  $\mathbf{B}_{\Pi SZ}$ ; but there are components of  $\mathbf{B}_{\Pi S}$  in  $X$ ,  $Y$ , and  $Z$ , depending on the

scattering angle. These are all generated from the incoming  $B_{\Pi Z}$  by tensor multiplication with  $\Xi_{\alpha\beta}$  in its second-rank tensor form.

Without loss of generality we concentrate on forward scattering in the rest of this section, so that<sup>20</sup>

$$\operatorname{Re}(a'_{\alpha\beta}) = \alpha'_{\alpha\beta}(f) + \zeta'_{\alpha\beta\gamma}(f)n'_\gamma + \alpha''_{\alpha\beta}(g) + \zeta''_{\alpha\beta\gamma}(g)n \quad (23)$$

$$\operatorname{Im}(a'_{\alpha\beta}) = -\alpha''_{\alpha\beta}(f) - \zeta''_{\alpha\beta\gamma}(f)n'_\gamma + \alpha'_{\alpha\beta}(g) + \zeta'_{\alpha\beta\gamma}(g)n_\gamma + \dots \quad (24)$$

where  $\alpha'_{\alpha\beta}$  and  $\alpha''_{\alpha\beta}$  are respectively the real and imaginary parts of the molecular polarizability tensor<sup>20</sup> and  $\zeta'_{\alpha\beta\gamma}$  and  $\zeta''_{\alpha\beta\gamma}$  are those of the zeta tensor defined by Barron.<sup>20</sup> In the forward direction, the process becomes one of antisymmetric spectral absorption, in which the incoming and outgoing  $B_{\Pi Z}$  and  $B_{\Pi SZ}$  define the absorption coefficient:

$$A^{(\Lambda)} \propto \frac{I_Z^{(s)}}{I_Z^{(0)}} \frac{B_{\Pi SZ}}{B_{\Pi Z}} = \Xi_Z \quad (25)$$

After ensemble averaging<sup>20</sup>

$$\langle \Xi_Z \rangle = \frac{\omega^4 \mu_0^2}{192\pi^2 R^2} (a_{\alpha\alpha} a_{\beta\beta}^* - \alpha_{\alpha\beta} \alpha_{\alpha\beta}^*) \quad (26)$$

and

$$B_{\Pi SZ} = \langle \Xi_Z \rangle B_{\Pi Z} \quad (27)$$

describes the process in terms of tensor invariants<sup>20</sup> of the molecular ensemble constituting the scattering volume.

#### IV. DISCUSSION

The historical development and experimental evidence for antisymmetric Rayleigh scattering has been reviewed in detail by Barron.<sup>20</sup> In this work and that of Knast and Kielich,<sup>7</sup> the magnetic nature of the phenomenon is implied indirectly, for example, through the fact that the process is described with the antisymmetric part of molecular property tensors such as the electric polarizability. Using the relation

$$\alpha''_{\alpha\beta} = \frac{1}{2} \varepsilon_{ijk} (\alpha''_{ij} - \alpha''_{ji}) \quad (28)$$

this tensor can be described as an axial vector,  $\alpha''_k$  (Refs. 21–25), which has the same symmetry as a magnetic dipole moment, and whose irreducible representations in various molecular point groups are the same as those of the magnetic dipole moment or angular momentum. Knast and Kielich,<sup>7</sup> in their Eq. (36), also point out that the vector  $\alpha''_k$  is negative to the motion reversal operator  $\hat{T}$ , and it follows<sup>21–25</sup> that it can form an interaction energy only with another  $\hat{T}$ -negative property, the antisymmetric conjugate product  $\mathbf{E} \times \mathbf{E}^*$  of Eq. (2) of this paper. This argument shows that  $\mathbf{E} \times \mathbf{E}^*$  must have the symmetry of a magnetic field, since  $\alpha''_k$  has the symmetry of a magnetic dipole moment. In fact, as discussed in the introduction,  $\mathbf{E} \times \mathbf{E}^*$  is proportional to the novel magnetic field  $\mathbf{B}_{\Pi}$ , the classical equivalent of the operator  $\hat{B}_\pi$  for each individual photon. Equation (13) of this paper now shows that antisymmetric scattering can be thought of as the induction by the scattered magnetic field  $\mathbf{B}_{\Pi S}$  of the magnetic dipole moment  $\langle \mathbf{m}_S(t) \rangle$  (which is the value at  $t$  of a time-correlation function), and this result can also be generalized in quantum field theory, or the theory of magneto-photonics. The Fourier transform of  $\langle \mathbf{m}_S(t) \rangle$  is a point on the spectrum of scattered light at the frequency  $\omega$ . The magnetic dipole moment  $\langle \mathbf{m}_S \rangle$  has the same irreducible representations in the appropriate point groups<sup>7</sup> as the antisymmetric polarizability considered by Knast and Kielich,<sup>7</sup> and  $\mathbf{B}_{\Pi S}(t)$  has the same symmetry as the antisymmetric part of the scattered intensity, denoted in tensor notation by  $I_{ij}^-$  by these authors.<sup>7</sup> It follows that the same conclusions arrived at by Knast and Kielich<sup>7</sup> for the properties of the antisymmetric polarizability hold for the novel magnetic dipole moment  $\langle \mathbf{m}_S(t) \rangle$ . For example,  $\langle \mathbf{m}_S(t) \rangle$  is nonzero only in the presence of a  $\hat{T}$ -negative influence, which in Eq. (13) is the magnetic field  $\mathbf{B}_{\Pi S}(t)$ .

Another conclusion that becomes immediately obvious in our magnetic interpretation of forward antisymmetric Rayleigh light scattering is that it involves circular polarization. The magnetic fields  $\mathbf{B}_{\Pi}$  and  $\mathbf{B}_{\Pi S}$  vanish if there is no degree of circular polarization, respectively in the incoming and scattered radiation. These findings are reinforced by the arguments, summarized in Section 3.5.3 of Ref. 20, for Rayleigh scattering in the near forward direction from refringent scattering theory. In this case, the degree of circular polarization is directly proportional to the pseudoscalar magnitude<sup>12</sup> of the scattered  $\mathbf{B}_{\Pi S}$ , which is the third Stokes parameter of the scattered radiation. Thus, in purely antisymmetric, near forward Rayleigh scattering, if the incident beam is completely circularly polarized, so is the scattered beam. This is summarized in our terms by Eq. (27), in which  $B_{\Pi Z}$  and  $B_{\Pi SZ}$  are both well defined, and in which the coefficient  $\langle \Xi_Z \rangle$  is a finite ensemble average. Clearly, if  $B_{\Pi Z}$  is zero (no degree of circular polarization in the incoming beam), then  $B_{\Pi SZ}$  is also zero,

because the molecular ensemble average  $\langle \mathbf{E}_Z \rangle$  is nonzero in general. Therefore, there is no near forward scattering.

## V. CONCLUSION

The phenomenon of antisymmetric light scattering has been interpreted in terms of the novel incident and scattered magnetostatic flux density vectors  $\mathbf{B}_\Pi$  and  $\mathbf{B}_{\Pi S}$ , respectively. This shows that antisymmetric scattering is a purely magneto-optic phenomenon, giving information on the nature of the scattered  $\mathbf{B}_{\Pi S}$  vector. In magneto-photonics, the vector  $\mathbf{B}_\Pi$  is replaced by the operator  $\hat{\mathbf{B}}_\Pi$ , and the appropriate quantum theory must be employed.

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## MANIFESTLY COVARIANT THEORY OF THE ELECTROMAGNETIC FIELD IN FREE SPACETIME, PART I: ELECTRIC AND MAGNETIC FIELDS AND MAXWELL'S EQUATIONS

### I. INTRODUCTION

It has recently been shown<sup>1-5</sup> that there exist longitudinal solutions of Maxwell's equations in free spacetime which are independent of the phase  $\phi$  of the traveling plane wave. These longitudinal electric and magnetic fields, denoted  $\mathbf{E}^{(3)}$  and  $\mathbf{B}^{(3)}$ , respectively, are consistent with the conclusion of quantum electrodynamics that there exist four photon polarizations in free spacetime, one timelike (00), two transverse spacelike (11) and (22), and one longitudinal spacelike (33).<sup>6,7</sup> However, the existence of four photon polarizations has to date been regarded<sup>7</sup> as being in conflict with the deduction that the photon can have only two helicities, +1 and -1. This in turn has led to the arbitrary assertion that only the two transverse spacelike polarizations (1) and (2) can be "physically meaningful" in free spacetime. The timelike (00) and longitudinal spacelike (33) are conventionally discarded as physically meaningless. This implies that the theory of the electromagnetic field in free spacetime loses manifest covariance.<sup>7</sup> This fundamental difficulty is well described by Ryder,<sup>7</sup> from whose Chapter 4 we quote the following: "the electromagnetic field, like any massless field, possesses only two independent components, but is covariantly described by a (potential) four vector  $A_\mu$ . In choosing two of these components as the physical ones, and thence quantizing them, we lose manifest covariance. Alternatively, if we wish to keep covariance, we have two redundant components."