

Acknowledgments

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THE PHOTON'S MAGNETOSTATIC FLUX QUANTUM: THE OPTICAL COTTON-MOUTON EFFECT

I. INTRODUCTION

The ability of circularly polarized electromagnetic radiation to produce anisotropy in magnetic permeability was first proposed by Piekara and Kielich,^{1,2} who systematically described light-induced anisotropy in material electric permittivity ($\Delta\epsilon$), magnetic permeability ($\Delta\mu$), and refractive index (Δn). In Ref. 1 for example, formulated in the pre-laser era, it was proposed that "On observe alors des changements de ϵ , μ , ou n , dus à l'action du champ polarisant." We are concerned in this paper with the formulation of an optical Cotton-Mouton effect, a relative of the optical Kerr effect first proposed by Buckingham³ and classified by Piekara and Kielich in their references. We define the novel optical Cotton-Mouton effect as a change in refractive index (linear dichroism) due to the novel, recently proposed, static magnetic field (\mathbf{B}_Π) of a circularly polarized electromagnetic plane wave.⁴⁻⁸ Piekara and Kielich^{1,2} described "saturation optique dans un champ optique." This effect later became known as the optical Kerr effect, or Buckingham effect.³

This paper is developed from the recent deduction⁴⁻⁸ that the photon carries a magnetostatic flux quantum, \hat{B}_Π , whose classical equivalent is a phase-independent magnetic field \mathbf{B}_Π generated in a circularly polarized light beam, an axial vector with the symmetry characteristics of a static magnetic flux density (tesla). The latter must be an axial vector positive to the parity inversion operator \hat{P} , and negative to the motion reversal operator \hat{T} (Ref. 9). The classical field \mathbf{B}_Π of the circularly polarized electromagnetic plane wave is a purely real quantity that is proportional to the antisymmetric (purely imaginary) part of the tensor $E_i E_j^*$, where E_i is the electric field strength of the wave in volts per meter. The scalar part of the tensor $E_i E_j^*$ is proportional to the phase-independent intensity of the plane wave in watts per meter squared, and we have shown elsewhere⁴⁻⁸ that the vector part of $E_i E_j^*$ (i.e., its antisymmetric part) is proportional to the phase-independent magnetic flux density \mathbf{B}_Π and vanishes if there is no degree of circular polarity. Furthermore, we have shown⁸ that \mathbf{B}_Π can be expressed in terms of the ubiquitous third Stokes parameter S_3 (Ref. 10) and therefore that phenomena such as circular dichroism and ellipticity are fundamentally magnetic.

The definition⁴⁻⁸ of the \hat{B}_Π operator per photon allows a wide range of novel optical/photonic phenomena to be forecasted straightforwardly, on the grounds that circularly polarized electromagnetic radiation can magne-

size. This conclusion is independent of the phase of the plane wave, and therefore independent of its angular frequency, ω (rad/s). It follows that the time average of the classical vector \mathbf{B}_Π is nonzero. It is emphasized that \mathbf{B}_Π is fundamentally different from the usual oscillating \mathbf{B} field of the plane wave¹⁰: \mathbf{B} vanishes when time averaged, because it is phase dependent, and has no component in the propagation axis Z of the wave. The vector \mathbf{B}_Π is directed exclusively in Z , and has no components in X and Y . By expressing the antisymmetric part of the tensor $E_i E_j^*$ as a vector product, $\mathbf{E} \times \mathbf{E}^*$ (refs. 4-8), it becomes clear that \mathbf{B}_Π is a relative of the Poynting vector,⁸⁻¹⁰ $\mathbf{N} = (\mathbf{E} \times \mathbf{B}^*)/\mu_0$, where μ_0 is the free space permeability. However, the polar vector \mathbf{N} is \hat{T} - and \hat{P} -negative, whereas the axial vector \mathbf{B}_Π is \hat{T} -negative, \hat{P} -positive,^{4-8,11} a critically important symmetry difference. Accordingly, \mathbf{N} is interpreted physically as a flux of energy density, and \mathbf{B}_Π as a flux of magnetic density. Remarkably, \mathbf{N} has been well known for many years, and \mathbf{B}_Π appears to be entirely novel.

The flux density vector \mathbf{B}_Π is clearly generated in vacuo (i.e., in free space), in direct analogy with \mathbf{N} . Both vectors \mathbf{N} and \mathbf{B}_Π are generated from solutions of Maxwell's classical equations through vector cross products of the usual, oscillating, phase-dependent \mathbf{E} and \mathbf{B} components of the electromagnetic plane wave solutions, cross products that multiply a vector with a complex conjugate vector, thus removing the phase dependence. For example, the complex conjugate (\mathbf{E}^*) of \mathbf{E} , a plane wave solution of Maxwell's equations, is also an allowed solution of Maxwell's equations, and the vector product of \mathbf{E} and \mathbf{E}^* , two allowed solutions, generates the purely imaginary conjugate product $\Pi^{(A)}$, which is proportional⁴⁻⁸ to \mathbf{B}_Π . Similarly, the vector product of \mathbf{E} and \mathbf{B}^* is proportional to \mathbf{N} . Therefore, although Maxwell's equations allow no direct, phase-independent solutions in free space, vector products of allowed solutions, such as \mathbf{N} and \mathbf{B}_Π , are physically meaningful phase-independent quantities whose time averages are nonzero.

It follows that \mathbf{B}_Π can interact with material to produce observable effects, again in direct analogy with \mathbf{N} . The scalar part of \mathbf{N} is the intensity I_0 , and the intensity (for example, of a laser) is clearly a free space quantity that affects and interacts with material. (For example, a sample is heated by intense light, light that travels through a vacuum.) Similarly, \mathbf{B}_Π is a free space magnetic flux density that can also affect material. For example, \mathbf{B}_Π forms a vector dot product with an electronic or nuclear magnetic dipole moment to give an interaction Hamiltonian (whose expectation value is an observable and measurable energy). This leads to the recently observed phenomenon of optical NMR¹² in which a circularly polarized laser shifts NMR resonances in new and unexpected ways, leading to useful new fingerprints for the analytical laboratory.¹³ These shifts were found experimentally to vanish in the uncertainty of measure-

ment when the laser's circular polarization was removed, strong evidence that they depend in an as yet incompletely understood manner on \mathbf{B}_Π . (There are as many, if not more, mechanisms involving \mathbf{B}_Π in optical NMR as there are involving the ordinary magnetostatic field in conventional NMR.)

It is easy to see that if circularly polarized light is simply regarded as an "optical magnet," there should be observable in one way or another all the well-known phenomena of conventionally produced magnetism,¹⁴ such as the Faraday, magnetic circular dichroic, Zeeman, Cotton-Mouton, Gerlach-Stern, Aharonov-Bohm, NMR, and ESR phenomena. Thus far, the \mathbf{B}_Π concept has been developed for the optical Zeeman effect,⁵ anomalous optical Zeeman effect,⁶ the optical Faraday effect,⁷ optical effects in Compton scattering,⁴ and the inverse Faraday effect,⁸ which is bulk magnetization by \mathbf{B}_Π of a circularly polarized laser. In general, whenever a magnetic can be used in physics, so can a circularly polarized laser, which generates \mathbf{B}_Π . The magnitude of \mathbf{B}_Π is approximately $10^{-7} I_0^{1/2}$ in tesla,⁴⁻⁸ so that an accurately circularly polarized laser of intensity 1.0 W cm^{-2} generates 10^{-5} T , about a tenth of the earth's mean magnetic field. Clearly, pulses of laser radiation of say, up to 10^{16} W m^{-2} , available in principle,¹⁵ generate a substantial 10 T over the duration of the laser pulse. (Normally incoherent radiation, such as daylight, produces no \mathbf{B}_Π , because there is no mean circular polarization; a linearly polarized laser, however, intense, produces no \mathbf{B}_Π , because such a laser always contains equal and opposite amounts of right and left circularly polarized light—right and left photons.)

In this paper, an example is given of the straightforward way in which the \mathbf{B}_Π vector can be used to anticipate the existence of a novel optical phenomenon—the optical Cotton-Mouton effect. Section II defines \mathbf{B}_Π in its classical limit in terms of fundamental constants, and brings out the precise analogy between \mathbf{B}_Π and an ordinary magnetostatic flux density, \mathbf{B}_0 . This allows the semiclassical theory¹⁰ of the Cotton-Mouton effect to be developed straightforwardly in terms of \mathbf{B}_Π in section III. The order of magnitude of the linear dichroism (or ellipticity) induced by \mathbf{B}_Π is estimated in Section IV.

II. DEFINITION OF THE CLASSICAL \mathbf{B}_Π OF A CIRCULARLY POLARIZED LASER

The classical vector \mathbf{B}_Π of a circularly polarized laser in free space is obtained straightforwardly⁴⁻⁸ by a consideration of the \hat{T} and \hat{P} symmetries of the conjugate product—the vector part of $E_i E_j^*$:

$$\Pi^{(A)} = \mathbf{E} \times \mathbf{E}^* = 2E_0^2 \mathbf{k} = 2E_0 c i \mathbf{B}_\Pi \quad (1)$$

Here the axial vector \mathbf{B}_{Π} is in tesla, is \hat{T} -negative and \hat{P} -positive, and is directed in the propagation axis of the laser. Thus, \mathbf{B}_{Π} has the necessary and sufficient characteristics to define a magnetic flux density vector. This simple derivation shows that circularly polarized radiation magnetizes material with which it comes into contact from free space. A relation between \mathbf{B}_{Π} and the Poynting vector \mathbf{N} is obtained straightforwardly from a consideration of¹⁰

$$\mathbf{B} = \frac{1}{c} \mathbf{n} \times \mathbf{E} \quad \mathbf{E} = -\frac{c}{n^2} \mathbf{n} \times \mathbf{B} \quad (2)$$

Here, \mathbf{n} is a \hat{T} - and \hat{P} -negative polar vector, whose scalar magnitude is the refractive index, and which is related to the classical wave vector of the laser by

$$\mathbf{k} = \frac{\omega}{c} \mathbf{n} \quad (3)$$

In free space, the scalar magnitude of \mathbf{n} is unity, and it follows that

$$\mathbf{N} = 2I_0 \mathbf{n} \quad (4)$$

where the magnitude of \mathbf{N} is defined through the scalar intensity of the laser in $\mathbf{W} \text{ m}^{-2}$:

$$I_0 = \epsilon_0 c E_0^2 \quad (5)$$

Here ϵ_0 is the vacuum permittivity in S.I. units and c is the speed of light. It follows that \mathbf{B}_{Π} is related to the square root of the Poynting vector:

$$\mathbf{B}_{\Pi} = B_0 \mathbf{k} = \frac{E_0}{c} \mathbf{k} = \left(\frac{I_0}{\epsilon_0 c^3} \right)^{1/2} \mathbf{k} = \left(\frac{|\mathbf{N}|}{2\epsilon_0 c^3} \right)^{1/2} \mathbf{k} \quad (6)$$

From these simple derivations it follows that the scalar part (or trace) of the tensor $E_i E_j^*$ is responsible for the Poynting vector's magnitude, and that the antisymmetric (vector) part of the tensor $E_i E_j^*$ is responsible for the novel phase-independent magnetic flux density \mathbf{B}_{Π} . In quantum field theory it has been shown elsewhere⁴⁻⁹ that \mathbf{B}_{Π} becomes a novel elementary magnetic field of the photon itself—an operator \hat{B}_{Π} .

III. APPLICATION TO THE OPTICAL COTTON-MOUTON EFFECT

Since \mathbf{B}_{Π} has all the characteristics of a magnetostatic flux density, it can be used to describe a variety of novel magneto-optic effects, an example of which is an optical Cotton-Mouton effect, developed in this section with a standard semiclassical approach. The optical Cotton-Mouton effect is the development of linear birefringence in a probe light beam propagating in axis Z through a suitable sample and linearly polarized at 45° to the direction of an applied pump laser in the X axis. The latter is circularly polarized and generates $B_{\Pi X}$. Elliptical polarization in the probe is produced by $B_{\Pi X}$ of the pump, which plays the role of the ordinary magnet of the original effect discovered by Cotton and Mouton¹⁶ in 1907. The pump's $B_{\Pi X}$ produces a phase difference in the two coherent resolved components of the probe, linearly polarized parallel and perpendicular, respectively, to the direction X of $B_{\Pi X}$ of the pump. This phase difference is¹⁰

$$\delta = \frac{\omega}{c} l (n_{\parallel} - n_{\perp}) \quad (7)$$

where n_{\parallel} and n_{\perp} are the refractive indices for light linearly polarized parallel and perpendicular to X . The resulting ellipticity is $\delta/2$.

At absorbing wavelengths, the two components n_{\parallel} and n_{\perp} are accompanied by two different absorption coefficients, signaling the presence of linear dichroism due to $B_{\Pi X}$ of the circularly polarized pump laser. There is a rotation of the major axis of the polarization ellipse of the probe laser because a difference in amplitude develops between two orthogonal resolved components for which no phase difference exists.¹⁰

Kielich and Piekara² have summarized the various theories of the standard Cotton-Mouton effect, under their classification scheme denoted "optical saturation in a magnetic field." In our case this magnetic field is $B_{\Pi X}$ of the circularly polarized pump laser in direction X . The novel \mathbf{B}_{Π} concept allows these theories to be adapted directly for the optical Cotton-Mouton effect suggested here. We have simply replaced an ordinary magnet with an optical magnetic, which is an intense circularly polarized pump laser, operable at any electromagnetic frequency, from infrared to X-ray regions. Notable theories include those of Raman and Krishnan,¹⁷ Piekara,^{18,19} Peterlin and Stuart,²⁰ Snelman,²¹ and the semiclassical approach at Buckingham and Pople.^{22,23} Kielich has developed the conventional Cotton-Mouton and related effects in several directions, for example, (1) the theory of the inverse Cotton-Mouton effect,²⁴ which he described as the induction of magnetic anisotropy by an intense laser

beam; (2) the theories in colloids of the inverse Cotton-Mouton effect²⁵ and the important but neglected Majorana effect²⁶ in colloids, liquid crystals, and polymers; and (3) general theories of magneto-optics.²⁷

These theories can now be recast to great advantage, in principle, using the \mathbf{B}_Π concept, or its equivalent for magneto-photonics, the operator \hat{B}_Π (Ref. 15) of the photon itself. As an example we take the semiclassical theory of Buckingham and Pople²³ given originally for the ordinary Kerr effect, and adapt it straightforwardly for the optical Cotton-Mouton effect by substituting \mathbf{B}_Π for the ordinary static electric field \mathbf{E}_0 of the Kerr effect, or the ordinary static \mathbf{B}_0 field of the standard Cotton-Mouton effect.¹⁰ In so doing it is convenient to follow the summary given by Barron¹⁰ for the Kerr effect and indicate the simple changes needed for the optical Cotton-Mouton effect along the way.

The starting point is the expression for probe ellipticity in Rayleigh refringent scattering theory¹⁰:

$$\eta = -\frac{1}{4}N\omega\mu_0c[\alpha'_{XX}(f) - \alpha'_{YY}(f)] \quad (8)$$

in terms of laboratory frame components of the real parts of the polarization tensor $\alpha'_{\alpha\beta}$ of a molecules of the sample. Here N is the number of molecules in the sample, ω is the angular frequency of the probe laser, μ_0 is the vacuum magnetic permeability, and l is the sample length in meters through which the probe passes. The \mathbf{B}_Π vector of the circularly polarized pump laser generates anisotropy in the sample because \mathbf{B}_Π interacts with the permanent and induced magnetic dipole moments in each molecule (or atom). The total magnetic dipole moment per molecule is, accordingly,

$$m_\alpha = m_{0\alpha} + \chi'_{\alpha\beta}B_{\Pi\beta} + \dots \quad (9)$$

where $m_{0\alpha}$ is the permanent molecular electronic magnetic dipole moment (if nonzero), and $\chi'_{\alpha\beta}$ is the real static susceptibility, a symmetric second-rank property tensor.¹⁰ The dynamic polarizability is perturbed by \mathbf{B}_Π of the pump laser as follows:

$$\alpha'_{\alpha\beta}(\mathbf{B}_\Pi) = \alpha'_{\alpha\beta}(0) + \alpha'_{\alpha\beta\gamma} + \alpha'_{\alpha\beta\gamma\delta}B_{\Pi\gamma}B_{\Pi\delta} + \dots \quad (10)$$

and in the evaluation of η in Eq. (8) an ensemble average is taken of the polarizability tensor components perturbed by \mathbf{B}_Π of the pump. In forming this ensemble average, an interaction potential energy is used of the type

$$V(\Omega) = -m_{0X}B_{\Pi X} - \frac{1}{2}\chi'_{XX}B_{\Pi X}^2 + \dots \quad (11)$$

From tensor invariant theory¹⁰ the ellipticity is finally obtained, in precise parallel with the theory of the Kerr effect, as

$$\begin{aligned} \eta = & -\frac{1}{120}\omega\mu_0c/NB_{\Pi X}^2 \left[\alpha'_{\alpha\beta\alpha\beta}(f) - \alpha'_{\alpha\alpha\beta\beta}(f) \right. \\ & + \frac{2}{kT} \left(3\alpha'_{\alpha\beta\alpha}(f)m_{0\beta} - \alpha'_{\alpha\alpha\beta}(f)m_{0\beta} \right) \\ & + \frac{1}{kT} \left(3\alpha'_{\alpha\beta}(f)\chi'_{\alpha\beta} - \alpha'_{\alpha\alpha}(f)\chi'_{\beta\beta} \right) \\ & \left. + \frac{1}{k^2T^2} \left(3\alpha'_{\alpha\beta}(f)m_{0\alpha}m_{0\beta} - \alpha'_{\alpha\alpha}(f)m_{0\beta}m_{0\beta} \right) \right] \end{aligned} \quad (12)$$

which is valid rigorously at transparent frequencies only.

IV. DISCUSSION

For simplicity we consider a sample that has no permanent magnetic dipole moment. For this sample the probably dominant term in Eq. (12) involves a product of the molecular polarizability and molecular susceptibility. The ellipticity developed in the probe is second order in $B_{\Pi X}$, or first order in the intensity I_0 of the pump laser. Accordingly, the sign of η should not be changed by switching the circular polarization of the pump from left to right, thus reversing \mathbf{B}_Π (Refs. 4–8). However, if the pump is linearly polarized, $B_{\Pi X}$ and thus η should be zero for all I_0 of the pump.

With these overall considerations and taking a sample molecular electric polarizability²⁷ of the order $10^{-40}\text{C}^2\text{m}^2\text{J}^{-1}$, a static molecular susceptibility of the order $10^{-24}\text{C}^2\text{m}^{-4}\text{J}^{-1}\text{S}^{-1}$, N of the order 10^{26} molecules m^{-3} , l of 1 m, ω about 10^{15} rad s^{-1} , and kT of 4.14×10^{-21} J, corresponding to 300 K, we obtain

$$\eta \doteq 10^{-2}B_{\Pi X}^2 \quad (13)$$

Therefore, for a pump laser delivering a $B_{\Pi X}$ pulse of 1.0 T, the ellipticity change is 0.01 rad, or 0.6°m^{-1} . As first discussed by Kielich,²⁵ this could be enhanced by up to six orders of magnitude in colloidal solution, or in suitable liquid crystals just above the isotropic to mesophase transition, i.e., in a state where the sample is still transparent to pump and probe lasers. The effect of the pump's \mathbf{B}_Π pulse can be picked up by a probe using highly developed contemporary timing technology, as in work on the rotation of the elliptical polarization ellipse by a circularly polarized, giant

ruby laser pump pulse.²⁸⁻³⁰ It therefore appears possible to observe the optical Cotton-Mouton effect as proposed in this work in terms of the novel \mathbf{B}_Π vector, whose photon equivalent is the \hat{B}_Π operator, the photon's magnetostatic flux density.

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THE PHOTON'S MAGNETOSTATIC FLUX QUANTUM: FORWARD-BACKWARD BIREFRINGENCE INDUCED BY A LASER

I. INTRODUCTION

When a magnetostatic flux density \mathbf{B}_S is applied to an initially isotropic chiral liquid, that liquid develops forward-backward birefringence, otherwise known as magneto-chiral birefringence and magneto-spatial dispersion. The refractive index in the direction (+Z) of forward propagation of a probe laser becomes different from that in the backward direction (-Z). The Kramers-Kronig theorem implies that the same happens to the power absorption coefficient. The presence of this effect in liquids has been proposed several times theoretically, but has never been detected experimentally. The effect appears to have been first proposed by Portugal and Burstein¹ in magnetic crystal symmetries, and was measured by Mankelov et al.² The theory was extended by Kielich and Zawodny³ to crystals with magnetic ordering. Working with liquids, Baranova and Zel'dovich⁴ described the refractive index change in circularly polarized probe radiation in terms of the dot product $\mathbf{B}_S \cdot \mathbf{\kappa}$, where $\mathbf{\kappa}$ is the classical wave vector of the probe, and implied the presence of forward-backward birefringence. The first detailed papers on the subject in chiral liquids are due to Woźniak and Zawodny,^{5,6} who defined the molecular point groups able to support the effect, and developed a theory based on electronic distortion and reorientation of the permanent molecular magnetic dipole moment, if nonvanishing. Wagnière and coworkers⁷⁻⁹ developed the theory of the effect for power absorption as well as refractive index, and Barron and Vrbancich¹⁰ contributed a comprehensive paper on forward-backward birefringence and dichroism in chiral liquids based on time-odd, complex, molecular property tensors. In this work, an unsuccessful attempt to measure the effect experimentally was reported briefly. Woźniak later developed the semiclassical theory of the effect in diamagnetic molecules