

THE OPTICAL FARADAY EFFECT AND OPTICAL MCD

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ABSTRACT

Based on the recent theoretical deduction of the photon's static magnetic field operator \hat{B}_{II} , the optical Faraday effect and optical MCD are proposed. It is shown that \hat{B}_{II} rotates the plane of a linearly polarized probe, an effect which is frequency dependent and which gives rise to optical magnetic circular dichroism (optical MCD), in which the conventional magnet is replaced by a \hat{B}_{II} generator, a circularly polarized laser generating a magneto-static field, each photon of which carries the quantum of flux \hat{B}_{II} .

INTRODUCTION

This paper continues a series of articles in which the consequences are developed of the recent deduction [1-6] that the photon generates a magnetic flux quantum, an operator

$$\hat{B}_{II} = B_0 \frac{\hat{J}}{\hbar} \quad (1)$$

Here B_0 is the scalar magnetic flux density amplitude of a beam of N photons (a circularly polarized generator laser), \hat{J} is the photon's angular momentum operator [7] whose eigenvalues are $\pm M_J \hbar$, where M_J is plus or minus one, and where \hbar is the reduced Planck constant. The operator \hat{B}_{II} changes sign with the circular polarity of the generator laser, is unlocalized in space, and has eigenvalues $\pm M_J B_0$ where B_0 is the laser's scalar flux density amplitude. Its classical equivalent is the axial vector \mathbf{B}_{II} , a novel magnetostatic flux density generated by circularly polarized electromagnetic plane waves [1-6]. The theoretical existence of \hat{B}_{II} is supported by the experimental evidence for light induced magnetic ef-

fects. The first to be described (in the sixties) was the inverse Faraday effect [8–14], and recently it has been shown [15,16] that NMR resonances are shifted in new and useful ways by the magnetizing effect of circularly polarized argon ion radiation at frequencies far from optical resonance (i.e. where the sample is transparent to the argon ion radiation and does not absorb it). Both these effects can be described in terms of the operator \hat{B}_{II} , or its classical equivalent, \mathbf{B}_{II} [1–6,17–23]. It has also been proposed [4–6] that there exists an optical Zeeman effect, in which \hat{B}_{II} splits singlet electric dipole transition frequencies in atoms; an anomalous optical Zeeman effect for atomic triplet states; and an optical Paschen-Back effect. It has also been proposed theoretically [2,3] that \hat{B}_{II} can be detected by examining spectrally a circularly polarized laser at visible frequencies reflected from an electron beam.

The existence of \hat{B}_{II} also implies that of an optical Faraday effect, in which it rotates the plane of a linearly polarized probe, an effect which is frequency dependent, and which gives rise therefore to optical magnetic circular dichroism (optical MCD spectroscopy). These effects are developed theoretically in this paper for atoms.

Mason [24] has given an interesting discussion of cause and effect in chirality, a discussion which includes some pertinent historical analysis of interest here. In 1846, Faraday showed experimentally [25] that a magnetostatic field induces optical activity in flint glass and other isotropic transparent media. In 1884, Pasteur [26] proposed on the basis of this result that the magnetic field represented a source of chirality. It is now known [27] that the magnetically induced optical activity in the Faraday effect has a fundamentally different symmetry [28–31] from that of natural optical activity. Nonetheless, it is interesting for our purpose in this paper to note, following Mason [24], that le Bel in 1874 [32] had independently proposed that circularly polarized radiation also provides a “chiral force” of the type envisioned by Pasteur emanating from the magnetostatic field used by Faraday. Enantio-differentiating photoreactions were indeed reported by Kuhn and Braun in 1929 [33], and it has been proposed repeatedly (e.g. Bonner [34]) that circularly polarized solar irradiation may be responsible for the well known preponderance in nature of one enantiomer over another. However, Mason [24] favours the universal and parity violating mechanism of the electroweak force as the origin of this dissymmetry because the electroweak force does not depend on time and location on the earth’s surface. (Natural solar radiation is only 0.1% circularly polarized, and equally and oppositely so at dawn and dusk [24].)

It is therefore interesting for our purpose to note that both circular polarity in light and the magnetostatic field have been proposed independently (by le Bel and Pasteur respectively) as sources of chirality. It had therefore been sensed more than one hundred years ago that these two concepts have something in common in their effect on material. Pershan et al. [2], in their first paper on the experimental demonstration of the magnetizing effect of circularly polarized giant ruby laser radiation, edged towards the concept of \hat{B}_{II} by describing the effect of the laser as being due to an effective magnetostatic field. In view of these indications, those by Kielich and co-workers [12,13], and by Atkins and Miller [14], the present author appears to have demonstrated conclusively [1–6] the fact that circularly polarized radiation generates the classical magnetostatic field \mathbf{B}_{II} whose equivalent in quantum field theory is the photon's flux quantum \hat{B}_{II} , a novel fundamental property of quantum field theory. As noted elsewhere, the concept of \hat{B}_{II} must be clearly and carefully distinguished from the well known ("standard IUPAC") oscillating \mathbf{B} field of the electromagnetic plane wave; the two are quite different [1–6] in nature.

We have therefore resolved [1–6] the conjectures of le Bel (1874) and Pasteur (1884) insofar as to show that circularly polarized light can indeed act in the same way as a magnetostatic field, a finding which implies the existence of several novel types of spectroscopy in circumstances where a conventionally applied magnet would be replaced by a circularly polarized laser.

In Section 1, the contemporary quantum theory of Faraday's effect of 1846 is developed succinctly for atoms, whereby it becomes relatively straightforward to show that the flux quantum \hat{B}_{II} must also generate Faraday's observation of optical activity in all material, inherently (structurally) chiral or otherwise. In our case, the magnet used by Faraday is replaced by a circularly polarized laser which generates \hat{B}_{II} , and is therefore referred to as the "generator laser".

Section 2 develops the frequency dependence of the optical Faraday effect in atoms through the properties of the magnetically (i.e. \hat{B}_{II}) perturbed antisymmetric polarizability of conventional contemporary Faraday effect theory [35], and therefore arrives at expressions in atoms for optical MCD.

Finally a discussion is given of possible experimental configurations and order of magnitudes of the expected optical Faraday effect in terms of the intensity in watts per unit area of the generator laser.

1. OPTICALLY INDUCED FARADAY ROTATION IN ATOMS

The contemporary quantum theory of the Faraday effect is based on the well known work by Serber [36] which was the precursor for the well known A, B and C terms [37]. This section develops the Serber theory for use with a flux quantum \hat{B}_{II} from the generator laser, and shows that the analogy between the optical and conventional Faraday effects is easily forged by using \hat{B}_{II} in place of the conventional magnetic field \mathbf{B}_0 from a magnet.

The starting point for the theory of the conventional Faraday effect in quantum mechanics is an equation for the rotation of the plane of linearly polarized radiation. This is derived [37] by a consideration of the effect of a magnetostatic field on the antisymmetric part of an atomic or molecular property tensor called the antisymmetric polarizability, α_{ij}'' :

$$\alpha_{ij}''(B_{ok}) = \alpha_{ij}''(\mathbf{O}) + \alpha_{ijk}'' B_{ok} \quad (2)$$

where α_{ijk}'' is a perturbation tensor of order three. Before embarking on detailed theoretical development it is instructive to consider the fundamental symmetries of these atomic property tensors (we restrict consideration in this paper to atoms) and to recall that the complex electronic electric polarizability, of which α_{ij}'' is the antisymmetric, imaginary component, is derived from time dependent perturbation theory within whose framework [37] α_{ij}'' is a product of two transition electric dipole moment operators. The perturbation tensor α_{ijk}'' in this context involves two of these and one transition magnetic dipole moment matrix element. This structure introduces frequency dependence into the conventional Faraday effect, as is well known, leading to MCD. Similarly, frequency dependence occurs in the optical Faraday effect, leading to optical MCD.

The fundamental symmetries considered are parity inversion (represented by the operator \hat{P}) and motion reversal (by the operator \hat{T}). In this context α_{ij}'' is negative to \hat{T} and positive to \hat{P} , while α_{ijk}'' is positive to both \hat{P} and \hat{T} . Therefore α_{ij}'' is finite only in the presence of a \hat{T} negative influence, such as \mathbf{B}_0 or \hat{B}_{II} , and this influence is mediated by α_{ijk}'' , which is finite for all atoms and molecules and is described by the ubiquitous B term [37]. The contemporary theory of Faraday's effect depends on a perturbation of a quantity α_{ij}'' which is itself the result of semi-classical time dependent second order perturbation theory. The basic reason for this is that the observable in the Faraday effect is an angle of rotation ($\Delta\theta$) (or alternatively a change in ellipticity $\Delta\eta$) which must be calculated from Maxwell's equations or Rayleigh refringent scattering theory [37].

Although the A term is closely related to the Zeeman effect [38] the observables of the two effects are quite different in nature, being traditionally an angle of rotation (Faraday effect) and a frequency shift in the visible frequency region (Zeeman effect). In consequence the latter can be described by an energy perturbation, while Faraday's effect needs perturbation of the antisymmetric polarizability, because energy does not appear directly in Maxwell's equations, which are needed to calculate refractive indices and therefrom $\Delta\theta$ and $\Delta\nu$.

With these considerations, the starting point for our development of the optical Faraday effect and optical MCD is the equation [37] for angle of rotation in the conventional quantum theory of the Faraday effect:

$$\Delta\theta \doteq \frac{1}{4} \omega\mu_0cl \left(\frac{N}{d_n} \right) B_{oZ} \sum_n \left(\alpha_{XYZ}'' - \alpha_{YXZ}'' + \frac{m_{nZ}}{kT} (\alpha_{XY}'' - \alpha_{YX}'') \right) \quad (3)$$

Here, $\Delta\theta$ is the angle of rotation of plane polarized probe radiation of angular frequency ω parallel to the conventionally generated magnetostatic flux density B_Z . The quantity N is the total number of molecules per unit volume in a set of degenerate quantum states of the atom, individually designated [37] ψ_n where d_n is the degeneracy and the sum is over all components of the degenerate set, with $N_d = Nd_n$. In eqn. (3), μ_0 is the vacuum permeability, c the speed of light, l the sample length, m_{nZ} the Z component of the atomic magnetic dipole moment in state n ; and kT is the thermal energy per atom. Recall that the atomic property tensors α_{ij}'' and α_{ijk}'' are derived from semi-classical, time dependent, perturbation theory and are frequency dependent in general, so that $\Delta\theta$ mapped over a frequency range has the appearance of a spectrum — the well known, conventional MCD spectrum.

Our task here is to incorporate the novel flux quantum \hat{B}_Π [1–6] into eqn. (3), and thus generate the optical Faraday effect and optical MCD.

In order to proceed, we note that α_{ij}'' and α_{ijk}'' as used in eqn. (3) are expectation values of the respective quantum mechanical operators $\hat{\alpha}_{ij}''$ and $\hat{\alpha}_{ijk}''$ in the same way that m_{ni} is an expectation value of the magnetic electronic dipole moment operator, \hat{m}_n . The quantity B_{oZ} is a classical magnetostatic vector component, and $\Delta\theta$ is an expectation value of the operator $\hat{\Delta\theta}$. It is convenient to transform the appropriate cartesian components of the operators $\hat{\alpha}_{ij}''$ and $\hat{\alpha}_{ijk}''$ into spherical form [37,39], using the Condon/Shortley phase convention:

$$\begin{aligned}\hat{\alpha}_{XY}'' &= -\frac{i}{2} \left[\sqrt{2} \hat{\alpha}_0^{1''} + (\hat{\alpha}_2^{2''} - \hat{\alpha}_{-2}^{2''}) \right] \\ \hat{\alpha}_{YX}'' &= \frac{i}{2} \left[\sqrt{2} \hat{\alpha}_0^{1''} - (\hat{\alpha}_2^{2''} - \hat{\alpha}_{-2}^{2''}) \right] \\ \hat{\alpha}_{XYZ}'' &= -\frac{i}{2} \left[\hat{\alpha}_0^{2''} + \frac{1}{\sqrt{6}} (\hat{\alpha}_2^{2''} + \hat{\alpha}_{-2}^{2''}) + \frac{1}{\sqrt{3}} (\hat{\alpha}_2^{3''} - \hat{\alpha}_{-2}^{3''}) \right] \\ \hat{\alpha}_{YXZ}'' &= \frac{i}{2} \left[\hat{\alpha}_0^{2''} - \frac{1}{\sqrt{6}} (\hat{\alpha}_2^{2''} + \hat{\alpha}_{-2}^{2''}) - \frac{1}{\sqrt{3}} (\hat{\alpha}_2^{3''} - \hat{\alpha}_{-2}^{3''}) \right]\end{aligned}\quad (4)$$

from which

$$\begin{aligned}\hat{\alpha}_{XY}'' - \hat{\alpha}_{YX}'' &= -\sqrt{2} i \hat{\alpha}_0^{1''} \\ \hat{\alpha}_{XYZ}'' - \hat{\alpha}_{YXZ}'' &= -i \hat{\alpha}_0^{2''}\end{aligned}\quad (5)$$

Both $\hat{\alpha}_{ij}''$ and $\hat{\alpha}_{ijk}''$ are purely imaginary in the appropriate spherical representation, indicating that the operators $i\hat{\alpha}_0^1$ and $i\hat{\alpha}_0^2$ are anti-hermitian, with purely imaginary eigenvalues [37]. (Note carefully, however, that the \hat{T} symmetry of $i\hat{\alpha}_0^1$ is negative, while that of $i\hat{\alpha}_0^2$ is positive. Both have positive \hat{P} symmetry.)

The next step in our development for atoms is to replace the vector component B_{oZ} of the conventional quantum theory by the quantum mechanical operator defined by eqn. (1), which has associated with it the angular momentum quantum number J . An immediate consequence of this replacement is the necessity to consider the magnetic field flux quantum using irreducible tensorial methods of angular momentum coupling theory in quantum mechanics [39–41]. In other words we are now considering a *quantized* photon angular momentum interacting with an atom, which in general contains, as usual, quantized orbital and spin electronic angular momentum. Without loss of generality we restrict consideration to atomic singlet states, in which there is a net orbital electronic angular momentum \hat{L} , but no net spin angular momentum \hat{S} .

With these considerations, eqn. (3) becomes, for the optical Faraday effect:

$$\Delta\theta = \langle JM_J; LM_L | \Delta\hat{\theta} | J'M'_J; L'M'_L \rangle = -\frac{1}{4} \omega\mu_0 c l N_n i \langle JM_J | \hat{B}_{\Pi} | J'M'_J \rangle \quad (6)$$

$$\times \left[\frac{\langle LM_L | \hat{m}_{n0}^1 | L'M'_L \rangle \langle LM_L | \hat{\alpha}_0^1 | L'M'_L \rangle}{kT} + \langle LM_L | \hat{\alpha}_0^2 | L'M'_L \rangle \right]$$

where we have used an *uncoupled* representation [39–41] to describe the net angular momentum generated during the interaction of photon and atom. This is justified through the fact that the azimuthal components of the operators \hat{m}_n and \hat{B}_{Π} are both well defined in the uncoupled representation [39–41], whereas in the coupled representation of the same problem the total angular momentum is defined but the individual azimuthal components are not. With these considerations, the expectation value of the angle of rotation in the optical Faraday effect is

$$\Delta\theta = \mp \frac{1}{4} \omega\mu_0 c l N_n i M_J B_0 \left[\frac{M_L^2 \langle L || \hat{m}_0^1 || L \rangle \langle L || \hat{\alpha}_0^1 || L \rangle}{L(L+1)(2L+1)kT} \right. \quad (7)$$

$$\left. + \frac{(3M_L^2 - L(L+1)) \langle L || \hat{\alpha}_0^2 || L \rangle}{(L(L+1)(2L+1)(2L+3)(2L-1))^{1/2}} \right]$$

where we have used the fact that the expectation value of the photon's \hat{B}_{Π} operator is $\pm M_J B_0$, positive for left circularly polarized radiation and negative for right circularly polarized radiation from the generator laser.

This is an expression for the angle of rotation induced in plane polarized probe radiation in a sample of atoms by a circularly polarized laser generating the flux quantum \hat{B}_{Π} . The observation of such a rotation would provide a test for the existence of \hat{B}_{Π} . Equation (7) is written out in terms of reduced matrix elements of dipole moment and atomic polarizability operators, matrix elements which are products of electric and magnetic dipole transition dipole moment matrix elements from time dependent perturbation theory [37]. These introduce frequency dependence into the angle of rotation of the optical Faraday effect.

The selection rules governing the various atomic property tensors are as follows:

$$\hat{\alpha}_0^1 : \Delta L = 0; \Delta M_L = 0; \quad (8)$$

$$\hat{\alpha}_0^2 : \Delta L = 0; \pm 2; \Delta M_L = 0;$$

the $\Delta L = \pm 1$ part is parity forbidden, as in magnetic dipole transitions.

2. OPTICAL MCD, FREQUENCY DEPENDENCE OF $\Delta\theta$

The origin of frequency dependence in the optical Faraday effect can be traced to semi-classical time dependent perturbation theory, which produces expressions for the polarizability components as given in the conventional theory of magnetic electronic optical activity [37]. These can be further developed as usual in terms of reduced matrix elements of electric and magnetic transition dipole moment operators. For a given generator laser intensity and frequency therefore the optical MCD spectrum is a plot of the \hat{B}_{II} induced angle of rotation $\Delta\theta$ against the frequency of the linearly polarized probe. Experimentally, this is built up in principle by replacing the conventional magnet of MCD apparatus by the circularly polarized generator laser.

DISCUSSION

It is interesting to note that using the concept of \hat{B}_{II} the theory of the optical Faraday effect can also be developed and understood simply by replacing the magnetic flux density vector component B_{oZ} wherever it occurs in the conventional theory of MCD by the quantity $\pm B_0 M_J$ where B_0 is the magnetic flux density amplitude of the generator laser and $\pm M_J$ the two possible azimuthal quantum numbers of the photon. Thus, the optical Faraday effect can be developed along the lines of the conventional counterpart in Serber's A, B, and C terms, a convenient description of which is given by Barron [37] in his equations (6.2.2) and (6.2.3).

It follows that the optical MCD spectrum, which would test for the existence of \hat{B}_{II} , would have the same characteristics as the conventional spectrum. If this were to be confirmed experimentally, it would be strong evidence for the photon's fundamental flux quantum \hat{B}_{II} introduced in refs. [1–6]. If the optical MCD spectrum were found to differ from the conventional MCD spectrum, it would indicate the presence of other mechanisms of magnetization by the generator laser, such as the induction of a magnetic dipole moment through [42–45]

$$m_i = {}^m\beta_{ijk}^{ee}(E_j E_k^* - E_k E_j^*) \equiv {}^m\beta_{ijk}^{ee} \Pi_{jk}^{(A)} \quad (9)$$

where ${}^m\beta_{ijk}^{ee}$ is a hyperpolarizability, and $\Pi_{jk}^{(A)}$ is the antisymmetric conjugate product of the generator laser [42–45].

Finally, for an order of magnitude estimation of the expected angle of rotation in a linearly polarized probe due to a generator laser of intensity $I_0 = 10^6$ watts per square metre, we use antisymmetric polarizabilities

computed *ab initio* by Manakov, Ovsiannikov and Kielich [13] in atomic $S = 1/2$ ground states as a guide to orders of magnitude. For example, in Cs at 9440 cm^{-1} we have $\hat{\alpha}_0^1 = 3.4 \times 10^{-39} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$. Focussing attention on the term in $\hat{\alpha}_0^1$ in eqn. (7), and using $N = 6 \times 10^{26}$ molecules per metre cubed, and the Bohr magneton for m_0^1 , we obtain for a generator laser delivering at 300 K:

$$B_0 \sim 10^{-7} I_0^{1/2} = 10^{-4} \text{ tesla} \quad (10)$$

an angle of rotation of 0.8 radians per metre, measurable easily with a spectropolarimeter.

This result should be proportional to the square root of the intensity, I_0 of the generator laser and inversely proportional to temperature. There is also a contribution from the rank three perturbing tensor of eqn. (7). These features would add to the evidence for the existence of \hat{B}_{II} already available from the inverse Faraday effect [8–14] and light induced NMR shifts [15, 16].

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