

PUMP LASER INDUCED NET ANGULAR MOMENTUM:
ANGULAR POLARISABILITY, INDUCED ELECTRIC POLARISATION,
AND THE INVERSE FARADAY EFFECT.

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Abstract

The conjugate product $\underline{\pi}$ of the oscillating electric field strengths of a pump laser imparts net, time independent, orbital angular momentum (\underline{J}) to an atom or molecule through a quantity, $\underline{d}_{\text{ys}}''$, which is defined as atomic or molecular electronic spin polarisability. This is shown to be responsible for net electric polarisation in chiral ensembles (a new effect) and magnetization in achiral and chiral ensembles (the inverse Faraday effect). Using a Nd:YAG pump laser, with an electric field strength of up to 10^9 volts m^{-1} ; the former is shown to be a strong effect in chiral ensembles, one which can be amplified still further by resonance with a probe laser in the visible / ultra violet. This is discussed in the context of P violation in achiral ensembles.

Introduction

The concept of conjugate product in the classical electromagnetic field was reviewed as early as the mid sixties by Ward (1) and Giordmaine (2). The conjugate product is derived essentially from the fact that the Maxwell equations give two possible complex conjugate solutions for the electric field strength and magnetic flux density of the classical electromagnetic field. An early example of the physical effect of a conjugate product was observed experimentally (3,4) as the

inverse Faraday effect, which is magnetization due to an intense laser field. This was explained theoretically by Atkins and Miller (5) in terms of the vector (cross) product between the two complex conjugates \underline{E}^+ and \underline{E}^- of the electric field strengths (volts m^{-1}) of the classical electromagnetic field. A good account of this has been given recently by Wagnière (6), who has shown the presence of magnetization due to the conjugate product between \underline{E}^+ and the magnetic flux density (\underline{B}) of the laser field, an effect called "inverse magnetochiral birefringence" (6) which occurs only in chiral ensembles.

In this paper we use symmetry and elementary perturbation theory to indicate the presence of ELECTRIC polarisation due to the conjugate product

$$\underline{\pi} = \underline{E}_L^- \times \underline{E}_L^+ = -\underline{E}_R^- \times \underline{E}_R^+ = -2E_0^2 i \underline{k} \quad (1)$$

This is shown to be a much bigger effect than inverse Faraday and magnetochiral magnetization, and to be frequency dependent, so that resonance is possible using a probe laser with apparatus similar to that used in the optical Kerr effect, i. e. a pump laser parallel to a tunable probe. It is mediated by a molecular property tensor component named "molecular electronic spin polarisability", a P positive, T negative, axial vector.

The product $\underline{\pi}$ of the oscillating electric fields of a pump laser, can be (7) generated in, for example, a neodymium doped yttrium aluminium garnet (Nd:YAG). Commercially available versions of this laser can deliver continuous, Q-switched, pulsed, or focussed electromagnetic radiation whose electric field strength can reach 10^9 volts m^{-1} (7). The parity inversion (P) and motion reversal (T) symmetries of $\underline{\pi}$ are respectively + and -, and in this paper it is shown that it interacts with an axial vector quantity $d_{1xy}'' - d_{1yx}''$, the atomic or molecular electronic spin polarisability, which has the same P and T symmetries. In symmetry terms, the product $-i d_{sz}'' \pi_z$ appearing in the interaction hamiltonian between d_{1s}'' and $\underline{\pi}$ is closely analogous with that between static magnetic flux density

(B), and electronic orbital (J) and spin (s) angular momenta, which define the molecular magnetic dipole moment

$$\underline{m} = \gamma_e (\underline{J} + 2.002 \underline{s}) - \quad (2)$$

where γ_e is the magnetogyric ratio.

The axial vector $\underline{\pi}$ produces (8-10) optical activity and spectral splitting analogous to the Faraday and Zeeman effects produced by B (11). It also produces an atomic or molecular net orbital angular momentum, J, which induces a net magnetic moment through the magnetogyric ratio (10), and couples to electronic and nuclear spins. This is responsible for the experimentally observed (3,4) inverse Faraday effect. The net angular momentum J is involved in principle with laser induced spin orbit coupling both with the electronic and nuclear spins, and in fine spectral detail which would provide detailed information on \underline{d}_{1s}'' , a fundamental atomic or molecular quantity. These frequency dependent spectral effects appear to have remained unobserved, apparently because the known (magnetization) effects of laser conjugate products are small and difficult to measure. Advances in pump laser technology (7) are rapidly making such spectroscopies feasible. For the new, MUCH BIGGER, $\underline{\pi}$ -induced electric polarisation introduced in this paper, the spectral observation of fine detail in chiral samples appears to be firmly within the grasp of contemporary laser spectroscopies. This new effect is due fundamentally to the fact that $\underline{\pi}$ also induces a net ELECTRIC molecular dipole moment through a parity odd dynamic molecular property tensor \underline{X}_{ij} which is defined here with elementary perturbation theory, and which is supported in chiral molecular ensembles. This defines the tensor \underline{X}_{ij} as a product of matrix elements of a transition electric dipole moment with the transition spin polarisability matrix elements. It multiplies the conjugate product $\underline{\pi}_j$ to give an induced electric dipole moment in chiral ensembles. The

effect is much bigger, in consequence, than magnetization of the type discussed by Atkins and Miller (5) and Wagnière (9).

In section 1, the source of \underline{J} , the conjugate product, is defined for a left and right circularly polarised Nd:YAG pump laser, and its physical origin discussed in terms of the helical pattern of the tip of an electric field vector of the laser as it propagates with photon momentum \underline{k} . Section 2 discusses the ways in which $\underline{\pi}_j$ can interact with a polarisable atom or molecule through the vector quantity \underline{X}_{ij} , and the molecular electronic spin polarisability vector. Section 3 applies elementary perturbation theory to find the quantum mechanical origin and nature of \underline{X}_{ij} and makes an analogy between the new effect of this paper and the closely related inverse Faraday (3-6) and inverse magneto chiral birefringence effects (9). Section 4 discusses $\underline{\pi}$ induced electric polarisation in some interesting contexts, including that of P violating electroweak interactions in achiral ensembles, using the strength of the new effect as a possible measure of P violating transitions, and thus leading towards the evolution of P violating spectroscopies mediated by the neutral intermediate vector boson recently observed experimentally in the well known CERN experiment (12).

1. Definition of the Conjugate Product $\underline{\pi}$.

The conjugate product $\underline{\pi}$ is defined by

$$\underline{\pi} = \underline{E}_L^+ \times \underline{E}_L^- = - \underline{E}_R^+ \times \underline{E}_R^- = 2E_0^2 i \underline{k} - (3)$$

where L and R respectively denote left and right circular polarisation of a pump Nd:YAG laser propagating in the Z axis of the laboratory frame (X, Y, Z). Here \underline{k} is a unit vector in Z, and i denotes the square root of minus one. The product $\underline{\pi}$ is therefore purely complex, and reverses sign with circular polarisation of the pump. For a given circular polarisation, it reverses sign with the direction of propagation of the laser, and is therefore odd to motion reversal symmetry T (11, 13, 14). It is

the product of two parity odd electric field strengths, and is therefore even to parity inversion P. Therefore its P and T symmetries are the same as those (15-17) of static magnetic flux density \underline{B}_S . Using the third principle (18-22) of group theoretical statistical mechanics, the influence $\underline{\pi}$ makes possible the existence of ensemble averages which include the symmetry of $\underline{\pi}$. In the point group of achiral ensembles, this symmetry is $D_2^{(1)}(-)$ (21), and in the point group of chiral ensembles it is $D_2^{(1)}(-)$ (22,23). The symmetry of magnetisation is $D_2^{(1)}(-)$ and that of electric polarisation is $D_u^{(1)}(+)$. Therefore, $\underline{\pi}$ can induce magnetization both in achiral and chiral ensembles through a mediating P positive, T positive molecular property tensor (the inverse Faraday effect). The third principle also shows that $\underline{\pi}$ -induced electric polarisation, on the other hand, can be sustained only in a chiral ensemble, and must be mediated by a T negative, P negative molecular property tensor.

Note that theories (1,2,5,9) of conjugate product effects include the realisation that conjugate products have the property

$$\underline{E}^- \times \underline{E}^+ = - \underline{E}^+ \times \underline{E}^- \quad - (4)$$

and in consequence, it is necessary to define a specific order of multiplication of the conjugates \underline{E}^- and \underline{E}^+ . Physically, this has to do with the fact that the pump laser propagates in a specific direction with a specific "spin", i.e. a specific circular polarisation, so that it has right or left hand screw symmetry. Clearly, the product

$$\underline{\pi}_\omega = \frac{1}{2} \left(\underline{E}^- \times \underline{E}^+ + \underline{E}^+ \times \underline{E}^- \right) - (5)$$

averages to zero, and to obtain either magnetization or polarisation due to laser conjugate products, a definition such (3) must be used.

2. Definition of the Molecular Spin Polarisability.

The energy of interaction of the pump laser and a molecule of an ensemble is expanded in a Taylor series about the energy in the absence of the field

$$W(\underline{E}_o) = W_o + E_{od} \left(\frac{\partial W}{\partial E_{od}} \right)_o + \frac{1}{2!} E_{od} E_{op} \left(\frac{\partial^2 W}{\partial E_{od} \partial E_{op}} \right)_o + \dots \quad (6)$$

Here, the subscript 0 denotes that the electric field strength of the pump laser is taken at the molecular origin (11). Eqn. (6) can be rewritten as

$$W(\underline{E}_o) = W_o - E_{od} \mu_d - \frac{1}{2!} E_{od} E_{op} \alpha_{1d\beta} - \dots \quad (7)$$

with the definitions

$$\mu_d \equiv - \left(\frac{\partial W}{\partial E_{od}} \right)_o ; \quad \alpha_{1d\beta} = - \left(\frac{\partial^2 W}{\partial E_{od} \partial E_{op}} \right)_o \quad (8)$$

for the laser induced dipole moment μ_d and polarisability $\alpha_{1d\beta}$. We are interested in the electronic polarisability due to π . This polarisability describes the distortion of the molecular electronic charge distribution due to the conjugate product of the pump laser, which is time independent from eqn (3). This defines the interaction hamiltonian

$$\Delta H = - \frac{1}{2} (E_d \times E_p) \alpha_{1\gamma} \quad (9)$$

Writing the axial conjugate product vector $\underline{\pi}$ as a second rank tensor

$$\pi_{\alpha\beta} = \begin{bmatrix} 0 & 2E_0^2 & 0 \\ -2E_0^2 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad - (10)$$

using the purely mathematical relation

$$A_i = \epsilon_{ijk} B_{jk} \quad - (11)$$

where ϵ_{ijk} is the Levi Civita symbol, we find that the hamiltonian (9) is a real, scalar quantity obtained from the tensor contraction of two second rank antisymmetric tensors and $d_{1\alpha\beta}$. In order to obtain a purely real interaction hamiltonian, the latter must be a complex rank two tensor, defined by

$$d_{1\alpha\beta} = d'_{1\alpha\beta} - i d''_{1\alpha\beta} \quad - (12)$$

Using the definition of tensor contraction:

$$\begin{aligned} A_{ij} B_{ij} &= A_{xx} B_{xx} + A_{xy} B_{xy} + A_{xz} B_{xz} \\ &+ A_{yx} B_{yx} + A_{yy} B_{yy} + A_{yz} B_{yz} \\ &+ A_{zx} B_{zx} + A_{zy} B_{zy} + A_{zz} B_{zz} \end{aligned} \quad - (13)$$

we obtain the real interaction hamiltonian

$$\Delta H = -E_0^2 (d''_{1xy} - d''_{1yx}) \quad - (14)$$

Using the tensor relation (11) this can be written, finally, as

$$\begin{aligned} \Delta H &= -\frac{1}{2} \pi_z (d'_{1sz} - i d''_{1sz}) \quad - (15) \\ &\equiv -\frac{1}{2} \pi_z d_{1sz} \end{aligned}$$

where d_{1s}'' is defined as the molecular electronic spin polarisability vector, a rank one axial vector

$$d_{1si}'' = \epsilon_{ijk} d_{1sjk}'' \quad - (16)$$

The individual components of this vector are imaginary components of the complex dynamic molecular electronic polarisability, defined (11) by

$$\begin{aligned} d_{1dp}' &= \frac{2}{\hbar} \sum_{j \neq n} \frac{\omega_{jn}}{\omega_{jn}^2 - \omega^2} \operatorname{Re} \left(\langle n | \mu_d | j \rangle \langle j | \mu_p | n \rangle \right) \\ &= d_{1pd}' \end{aligned} \quad - (17)$$

and

$$\begin{aligned} d_{1dp}'' &= -\frac{2}{\hbar} \sum_{j \neq n} \frac{\omega_{jn}}{\omega_{jn}^2 - \omega^2} \operatorname{Im} \left(\langle n | \mu_d | j \rangle \langle j | \mu_p | n \rangle \right) \\ &= -d_{1pd}'' \end{aligned} \quad - (18)$$

in the laboratory fixed frame (X, Y, Z). The semi-classical theory of molecular property tensors (11) shows that these are time odd, parity even, and are negative to interchange of subscripts X and Y

$$d_{1xy}'' = -d_{1yx}'' \quad - (19)$$

In eqns (17) and (18), ω_{jn} is the transition frequency in radians s^{-1}

$$\omega_{jn} = \omega_j - \omega_n \quad - (20)$$

between quantum states j and n, \hbar is the reduced Planck constant, and μ_d and μ_p are transition electric dipole moments.

Note that this definition is at transparent frequencies, and when measuring radiation from a probe laser with angular frequency ω is tuned to ω_{jn} , i.e.

$$\omega \doteq \omega_{jn} \quad - (21)$$

The spin polarisability \underline{d}_{1s}'' can be amplified greatly. In this condition, semi-classical theory (11) uses

$$\begin{aligned} (\omega_{jn}^2 - \omega^2)^{-1} &\rightarrow \frac{(\omega_{jn}^2 - \omega^2) + i\omega\Gamma_j}{(\omega_{jn}^2 - \omega^2)^2 + \omega^2\Gamma_j^2} \\ &\equiv f + ig \quad \text{--- (22)} \end{aligned}$$

where f and g are the dispersion and absorption lineshape functions.

Note that the hamiltonian (15) is the product of two axial vectors (rank one tensors), each of which is positive to P and negative to T. Its symmetry characteristics are therefore precisely analogous to the interaction hamiltonian

$$\Delta H_2 = -B_{oz} m_z \quad \text{--- (23)}$$

between static magnetic flux density (B_{oz}) and the molecular magnetic dipole moment (m).

Perturbation Theory.

The product $\underline{\pi}$ is clearly time independent, but is generated by a time dependent electromagnetic field. In this section, we use the time dependent Schrodinger equation to define the T and P negative molecular property tensor X_{ij} responsible for the electric dipole moment (μ_i) induced by π_j :

$$\mu_i = X_{ij} \pi_j + \dots \quad \text{--- (24)}$$

The time dependent Schrodinger equation is

$$\left(i\hbar \frac{\partial}{\partial t} - H \right) \psi_n' = V \psi_n' \quad \text{--- (25)}$$

with the perturbed wavefunction (11)

$$\psi_n' = \left[\psi_n^{(0)} + \sum_{j \neq n} \frac{\langle j | d_{1s\beta}'' | n \rangle E_0^2 \psi_j^{(0)}}{\hbar \omega_{jn}} \right] e^{-i\omega_n t} \quad (26)$$

for a transition from state n to j at the transition frequency ω_{jn} . The unperturbed hamiltonian is

$$H = \hbar \omega_{jn} \quad (27)$$

and the interaction hamiltonian is

$$\Delta H = -d_{1s\beta}'' E_0^2 \quad (28)$$

Elementary perturbation theory (11) then expresses the tensor in the form

$$X_{d\beta} = \frac{2}{\hbar} \sum_{j \neq n} \frac{1}{\omega_{jn}} \langle n | \mu_{\alpha} | j \rangle \langle j | d_{1s\beta}'' | n \rangle \quad (29)$$

where d_{1s}'' is given by (17) and (18).

Note that this is T and P negative, as required by symmetry, and for a spin polarisability of the order $10^{-38} \text{ J}^{-1} \text{ C}^2 \text{ m}^2$; a dipole moment of the order 10^{-30} C m ; for $\hbar = 10^{-34} \text{ J s}$, and for a transition frequency ω_{jn} of 10^{15} radians s^{-1} ; its order of magnitude is $10^{-51} \text{ C}^3 \text{ m}^3 \text{ J}^{-2}$, about the same as that of the molecular first electric hyperpolarisability (χ_{ijk}). (For example, the experimentally observed hyperpolarisability of methane is about $10^{-51} \text{ C}^3 \text{ m}^3 \text{ J}^{-2}$ (24)). However, the tensor X_{ij} differs from the customary χ_{ijk} because the latter is observable both in achiral and chiral ensembles, whereas X_{ij} appears only in chiral ensembles in the absence of tiny P violating electroweak interactions. Principle three of group theoretical statistical mechanics (18-22) explains this in terms of the one to one relation between the symmetries of cause (external influence) and effect (the experimental observable). If the external influence is a static electric field strength, and

he observable is the ensemble averaged electric dipole moment, with the same negative P and positive T symmetries, the effect is observable in achiral and chiral ensembles and the mediating tensors (the electric polarisability, hyperpolarisability, and so on) are finite both in chiral and achiral ensembles. However, if the external influence is $\underline{\pi}$ and the observable is \underline{m} , with opposite P symmetries, the effect is sustained only in chiral ensembles, and mediating P and T negative tensors such as X_{ij} vanish in achiral ensembles in the absence of P violation. This illustrates the simplicity and usefulness of principle three.

To express the inverse Faraday effect in terms of \underline{d}_{15}'' , the magnetic dipole moment induced by $\underline{\pi}$ is expressed as

$$m_i = \gamma_{ij} \pi_j + \dots \quad (30)$$

and perturbation theory gives

$$\gamma_{df} = -\frac{2i}{\hbar} \sum_{j \neq n} \frac{\langle n | m_d | j \rangle \langle j | d_{15p}'' | n \rangle}{\omega_{jn}} \quad (31)$$

This result is similar to eqn. (18) of Wagniere (6) for the paramagnetic contribution to the inverse Faraday effect, where the equivalent of spin polarisability is expressed, after isotropic averaging, through the vector product of transition electric dipole moments.

Eqns. (29) and (31) compare directly the new effect of this paper, and the inverse Faraday effect. The latter occurs in all ensembles by principle three because γ_{ij} mediates the influence π_j and the observable m_i , both P positive, T negative quantities. In quantum mechanics, m_i is also purely complex, so that γ_{ij} in eqn. (31) must be purely real, explaining the appearance of the T positive real rather than the negative imaginary part of the spin polarisability in eqn. (31).

The inverse Faraday effect, and its close relative, inverse magnetochiral birefringence (6), are however,

relatively much smaller than the effect mediated by the tensor of eqn (29), which is of the order of the molecular electric first hyperpolarisability. Wagniere has estimated that magnetization due to the inverse Faraday effect is of the order 10^{-6} G for a Nd:YAG pump delivering 10^8 W cm^{-2} . That of inverse magnetochiral birefringence is about 10^{-8} G, equivalent to a magnetic induction of about 10^{-11} T. Both paramagnetic magnetization effects depend on free spins being available, and are small in diamagnetic materials. Most molecules without unpaired electron spins are diamagnetic.

In contrast, the new electric polarisation effect introduced here appears to have many interesting consequences.

4. Discussion

The mediating tensor χ_{ij} can be amplified considerably by resonance, when the probe laser (or a frequency component of broad band probe radiation) satisfies the resonance condition

$$\omega \doteq \omega_{jn} \quad \text{--- (32)}$$

so that the denominators in (17) and (18) become very small. Therefore by tuning the probe to ω_{jn} and using intense pulses of the pump, the magnitude of the induced electric dipole moment in a chiral ensemble is increased by orders of magnitude. This technique combines probe resonance with the effect of the time independent π of the pump, and is therefore similar in concept to nuclear magnetic resonance, where the time independent influence is static magnetic flux density, and the resonating electromagnetic probe is at MHz/GHz frequencies, rather than visible/ultra violet radiation as in the new effect introduced here.

Further, interesting, effects appear to be possible by tuning both the probe and the pump to the same, transition, frequency ω_{jn} . This is particularly so when attempting to amplify the minute absorptions due to electroweak interactions

mediated by the neutral vector boson (25). These violate P symmetry and allow u to u and g to g transitions which violate Laporte's Rule (26). In consequence, there is a very small X_{ij} in ACHIRAL ensembles due to these elementary electroweak forces. It may be possible to amplify the P violating X_{ij} at a resonance frequency ω_{in} of a Laporte disallowed transition in an achiral atomic or molecular ensemble by tuning both the pump and probe to this frequency and using the full power of the pump in pulse trains.

We have shown in this paper that the conjugate product of a pump laser, delivering pulses of great power, induces in chiral ensembles a time independent electric dipole moment μ_i through the tensor X_{ij} , with order of magnitude and units of molecular first electric hyperpolarisability. Therefore, it can be used to investigate the general electric polarisation properties of chiral ensembles, using the laser instead of electrodes. For example, in non-linear dielectric relaxation (27) the strong electric field can be replaced and heating effects removed by pulsing the pump laser. The probe in this case is an alternating electric field to measure dielectric relaxation. In another context, the equivalent of Stark splitting is possible using Π instead of a static electric field, with the availability in pulse trains of electric field strengths of up to 10^9 volts m^{-1} . Stark splitting in the microwave could therefore be observed in detail with Π from a Nd:YAG laser, and it has been shown elsewhere (28-31) that Π induces circular birefringence, for example Faraday A, B, and C terms and accompanying Zeeman splitting in achiral and chiral ensembles, and also produces forward backward birefringence under appropriate conditions (32).

The effect introduced here must be distinguished carefully from the optical Kerr effect, in which ellipticity is induced in the probe by the pump. This originates in the perturbation of the dynamic polarisability by the square of the time dependent electric field strength of the pump, and occurs in achiral and achiral ensembles. The Kerr effect due to a static

electric field is neither a circular nor a forward backward asymmetry, the refractive index becomes different in mutually orthogonal axes of the laboratory frame. In other words it is linear dichroism and birefringence due to the static electric field \underline{E} applied perpendicular to the probe. In analogy the optical Kerr effect is also linear dichroism, and in contrast the effect due to $\underline{\pi}$ introduced here is accompanied by both circular (29) and forward backward (32) birefringence and dichroism.

Finally in this paper, we note that the effect of $\underline{\pi}$ on the molecular dynamics of an S enantiomer of bromochlorofluoromethane has been computer simulated using classical equivalents of the tensor X_{ij} , showing pronounced effects (33) on the orientational and rotational velocity time correlation functions, respectively Fourier transforms of the frequency dependence of dielectric loss in the MHz / GHz range, and of the far infra red frequency dependence in the GHz / THz range, up to about 200 cm^{-1} in the far infra red. The theory of this paper anticipates such bandshape effects through the f and g functions of eqns (18) and (22).

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