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

by
M. W. Evans,
Theory Center,
Cornell University,
Ithaca,
NY 14853.

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Abstract
The conjugate product $\mathbf{\Theta}$ of the oscillating electric field strengths of a pump laser [g per net, time independent, orbital angular momentum $l$ to an atom or molecule through a quantity, $\mathbf{\Theta}$, 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 magnetisation in chiral and chiral ensembles (the inverse Faraday effect). Using a Nd:YAG pump laser, with an electric field strength of up to $10^6$ volts m$^{-1}$, the former is shown to be a strong effect in chiral assemblies, 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 Giromini (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
In this paper we use symmetry and elementary perturbation theory to indicate the presence of electric polarization due to the conjugate product
\[ \Pi = \varepsilon_{\alpha} \times \mathbf{E} \cdot \mathbf{E} = -\varepsilon_{\alpha} \times \mathbf{E} \cdot \mathbf{E} = -\varepsilon_{\alpha} \cdot \mathbf{E} \times \mathbf{E} \]

This is shown to be a much bigger effect than inverse Faraday and magnetoelectric 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 polarizability," a positive, T negative, axial vector.

The product \( \mathbf{P} \) of the oscillating electric field of a pump laser can be \( \mathbf{E} \) generated in, for example, a microwave-doped yttrium aluminum garnet (YAG). Commercially available versions of this laser can deliver continuous, Q-switched, pulsed, or focused electromagnetic radiation whose electric field strength can reach \( 10^8 \) volts m\(^{-1}\) (1). The parity inversion \( \mathbf{P} \) and monochromatic \( \mathbf{P} \) symmetries of \( \mathbf{P} \) are, respectively, \( P \) and \( T \), and this paper is to show that it interacts with an axial vector quantity \( \mathbf{L} \), the atomic or molecular electronic spin polarizability, which has the same \( P \) and \( T \) symmetries. In symmetry terms, the product \( \mathbf{P} \cdot \mathbf{L} \) appearing in the interaction Hamiltonian between \( \mathbf{P} \) and \( \mathbf{L} \) is closely analogous with that between static magnetic flux density.
\[ m = Y_e \left( J + 2 \cdot 0.02 \cdot 2 \cdot \frac{L}{S} \right) - (2) \]

where \( Y_e \) is the magneto optic ratio.

The axial vector \( \mathbf{\text{A}} \) produces (8.10) optical activity and spectral splitting analogous to the Faraday and Zeman effects produced by \( \mathbf{B} \) (11). It also produces an atomic or molecular net orbital angular momentum, \( J \), which induces a net magnetic moment through the magneto optic 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 \( \mathbf{A} \), 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 (17) are rapidly making such spectroscopies feasible. For the new, MUCH BIGGER, \( \mathbf{A} \)-induced electric polarization 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 \( \mathbf{A} \) also induces a net ELECTRIC molecular dipole moment through a parity odd dynamic molecular property tensor \( X_{ij} \), which is defined here with elementary perturbation theory, and which is supported in chiral molecular ensembles. This defines the tensor \( X_{ij} \) as a product of matrix elements of a transition electric dipole moment with the transition spin polarization matrix elements. It multiplies the conjugate product \( \mathbf{A} \) 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 (3) and Magnéniere (9).

In Section 1, the source of $\mathbf{L}$, the conjugate product $\mathbf{L}$, is defined for a left and right circularly polarized Nd:YAG pump laser, and its physical origin is discussed in terms of the helical pattern of the tip of an electric field vector of the laser as it propagates with photon momentum $\mathbf{k}$. Section 2 discusses the ways in which $\mathbf{L}$ can interact with a polarizable atom or molecule through the vector quantity $\mathbf{X}$, and the molecular electronic spin-polarizability vector. Section 3 applies elementary perturbation theory to find the quantum mechanical origin and nature of $\mathbf{X}$ 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 $\mathbf{L}$ induced electric polarization in some interesting contexts, including that of $\mathbf{P}$ violating electronic interactions in achiral molecules, using the strength of the new effect as a possible measure of $\mathbf{P}$ violating transitions, and thus leading toward the evaluation of $\mathbf{P}$ 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 $\mathbf{L}$

The conjugate product $\mathbf{L}$ is defined by

$$\mathbf{L} = \mathbf{E}_L^+ \times \mathbf{E}_R^- - \mathbf{E}_R^+ \times \mathbf{E}_L^- = 2\mathbf{E}_L^2 - (2)$$

where $\mathbf{L}$ and $\mathbf{R}$ respectively denote left and right circular polarizations of a pump Nd:YAG laser propagating in the $z$ axis of the laboratory frame $(x, y, z)$, here $\mathbf{E}$ is a unit vector in $\mathbf{Z}$, and $\mathbf{L}$ denotes the square root of minus one. The product $\mathbf{L}$ is therefore purely complex, and reverses sign with circular polarization of the pump, for a given circular polarization, it reverses sign with the direction of propagation of the laser, and is therefore odd to motion reversal symmetry $T (11, 12, 14)$. It is
The product of two parity odd electric field strengths, and is therefore even to parity inversion P, therefore its P and T symmetric are the same as those (15-17) of static magnetic flux density B. Using the third principle (19-22) of group theoretical statistical mechanism, the influence of P makes possible the existence of ensemble averages which include the symmetry of 0\(^{P}\). In the point group of chiral ensembles, this symmetry is 0\(^{\pm}\)(3), 31\(^{\pm}\), and in the point group of chiral ensembles it is 0\(^{\pm}\)(20, 22, 23). The symmetry of magnetisation is 0\(^{P}\)\(^{\pm}\)(4) and that of electric polarisation is 0\(^{P}\)\(^{\pm}\)(6). Therefore, \(S\) can induce magnetisation both in chiral and chiral ensembles through a mediating P positive, T positive molecular property tensor (the inverse Faraday effect). The third principle also shows that 0\(^{P}\)\(^{\pm}\) induced electric polarisation, on the other hand, can only be sustained only in a chiral ensemble, and not by a T negative, P negative molecular property tensor.

Note that theories (1,2,5,6) of conjugate product effects include the realisation that conjugate products have the property

\[ E^- \times E^+ - E^+ \times E^- = (S) \]

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

\[ \Pi_{\omega} = \frac{1}{2} (E^- \times E^+ + E^+ \times E^-) \]

reduces to zero, and to obtain either magnetisation or polarisation due to laser conjugate products, a definition such as (5) must be made.
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

\[ W(\mathbf{E}_d) = W_0 + \mathbf{E}_{ad} \cdot \left( \frac{\partial W}{\partial \mathbf{E}_{ad}} \right) + \frac{1}{2!} \mathbf{E}_{ad} \cdot \mathbf{E}_p \cdot \left( \frac{\partial^2 W}{\partial \mathbf{E}_{ad} \partial \mathbf{E}_p} \right) + \ldots \]  

(6)

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

\[ W(\mathbf{E}_d) = W_0 - \mathbf{E}_{ad} \cdot \mathbf{M_d} - \frac{1}{2!} \mathbf{E}_{ad} \cdot \mathbf{E}_p \cdot d_{\mu\nu} \ldots \]  

(7)

with the definitions

\[ \mathbf{M_d} = -\left( \frac{\partial W}{\partial \mathbf{E}_{ad}} \right) \]  

(8)

\[ d_{\mu\nu} = -\left( \frac{\partial^2 W}{\partial \mathbf{E}_{ad} \partial \mathbf{E}_p} \right) \]  

(9)

for the laser induced dipole moment \( \mathbf{M}_d \) and polarisability \( d_{\mu\nu} \). We are interested in the electronic polarisability due to \( \mathbf{E}_p \).

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

\[ \Delta H = -\frac{1}{2} \left( \mathbf{E}_d \times \mathbf{E}_p \right) \cdot d_{\mu\nu} \]  

(10)
Writing the axial conjugate product vector $\mathbf{\Pi}$ as a second rank tensor

$$\mathbf{\Pi}_p = \begin{bmatrix} 0 & 2e_x & 2e_y \\ -2e_x & 0 & 0 \\ 2e_y & 0 & 0 \end{bmatrix}$$  \hspace{1cm} (10)

using the purely mathematical relation

$$A_i = \varepsilon_{ijk} B_j B_k$$  \hspace{1cm} (11)

where $\varepsilon_{ijk}$ is the Levi-Civita symbol, we find that the Hamiltonian (1) is a real, scalar quantity obtained from the tensor contraction of two second rank antisymmetric tensors $\mathbf{\Pi}_p$ and $\mathbf{\Pi}_q$. In order to obtain a purely real interaction Hamiltonian, the latter must be a complex rank two tensor, defined by

$$d_{1q} = d'_{1q} - i d''_{1q}$$  \hspace{1cm} (12)

Using the definition of tensor contractions

$$A_{ij} \beta^j = A_k b_{im} + A_x b_{mx} + A_y b_{my} + A_z b_{mz}$$

$$A_k B_k + A_x B_x + A_y B_y + A_z B_z$$

$$+ A_{mz} B_{mz} + A_{mx} B_{mx} + A_{my} B_{my}$$

we obtain the real interaction Hamiltonian

$$\Delta H = -\varepsilon_{ijk} (d_{1iq} - d_{1qk})$$  \hspace{1cm} (14)

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

$$\Delta H = \frac{1}{2} \varepsilon_{ijk} (d_{1iq} - d_{1qk})$$  \hspace{1cm} (15)

$$\Xi = -\frac{1}{2} \varepsilon_{ijk} d_{1qk}$$
where $\mathbf{d}_{\text{lec}}$ is defined as the molecular electronic spin polarizability vector, a rank one axial vector

$$\mathbf{d}_{\text{lec}} = \sum_j d_{ij} \mathbf{e}_i \mathbf{e}_j$$

(16)

The individual components of this vector are imaginary components of the complex dynamic molecular electronic polarizability, defined (11.7) by

$$d_{\text{lec}}^{ii} = \frac{\alpha}{\kappa} \sum_{j < \kappa} \frac{c_{ij}}{\mu_{ij}^{\text{el}} - \omega^2} \Re \left( < n \mid \mu_{ij} \mid \frac{1}{\kappa} > \right)$$

(17)

and

$$d_{\text{lec}}^{i\kappa} = -\frac{\alpha}{\kappa} \sum_{j < \kappa} \frac{c_{ij}}{\mu_{ij}^{\text{el}} - \omega^2} \Im \left( < n \mid \mu_{ij} \mid \frac{1}{\kappa} > \right)$$

(18)

in the laboratory fixed frame (x, y, z). The semi-classical theory of molecular property tensors (11.7) shows that these are time odd, parity even, and are negative to interchange of subscripts $\kappa$ and $\kappa'$. In (17) and (18), $\omega_{ij}$ is the transition frequency in radians$^{-1}$

$$\omega_{ij} = \omega_{ij}^{\text{el}}$$

(19)

in mgs $(17)$ and $(18)$, $\omega_{ij}^{\text{el}}$ is the transition frequency in mgs, $\omega_{ij}^{\text{el}}$ is the reduced Planck constant, and $\mu_{ij}^{\text{el}}$ and $\mu_{ij}^{\text{el}}$ are transition electric dipole moments.

Note that this definition is at transparent frequencies, and when measuring radiation from a probe laser with angular frequency $\omega$, it is tuned to $\omega_{ij}^{\text{el}}$, i.e.,

$$\omega = \omega_{ij}^{\text{el}}$$

(20)
The spin polarizability $\hat{\alpha}_{ij}$ can be amplified greatly. In this
motion, semi-classical theory (11) uses
\[ (\omega_0^2 - \omega^2)^{-1} \Rightarrow \frac{\omega_0^2}{(\omega_0^2 - \omega^2)^2} = \frac{1}{\omega_0^2} - \frac{1}{\omega^2} \]

where $\alpha$ and $\beta$ are the dispersion and absorption lineshape
functions.

Note that the hamiltonian $H$ is the product of two
tensors (rank one tensors), each of which is positive $P$ and
negative $T$. Its symmetry characteristics are therefore
closely analogous to the interaction hamiltonian
\[ \gamma \mathbf{u}_2 = \mathbf{B}_{2z} \mathbf{m}_z \]

when static magnetic flux density ($\mathbf{B}_{2z}$) and the molecular
magnetic dipole moment ($\mathbf{m}_z$).

Perturbation Theory.

The product $\mathbf{u}_2$ is clearly time-independent, but it is
in the form of a time-dependent electromagnetic field. In this
section, we use the time-dependent Schrödinger equation to define
$T$ and $P$ negative molecular property tensor $X_{ij}$
appropriate for the electric dipole moment $\mathbf{m}_z$ induced by
\[ \mathbf{u}_2 = X_{ij} \mathbf{u}_j + \ldots \quad (38) \]

in the time-dependent Schrödinger equation is
\[ \left( i \frac{\partial}{\partial t} - H \right) \psi_n' = \mathbf{V} \psi_n' \quad (39) \]
with the perturbed wavefunction \( \psi_n' \):
\[
\psi_n' = \left[ \psi_n + \epsilon \sum_j \langle \psi_n | \hat{H}_n | \phi_j \rangle \langle \phi_j | \hat{H}_n | \psi_n \rangle \right] e^{-i \omega \epsilon}
\]
for a transition from state \( n \) to \( j \) at the transition frequency \( \omega_n \). The unperturbed Hamiltonian is
\[
\hat{H} = \epsilon \omega \hat{J}_n
\]
and the interaction Hamiltonian is
\[
\hat{H}_n = - \hat{d}_{n\gamma} \hat{E} \hat{J}_n
\]
Elementary perturbation theory (11) then increases the tensor in the form
\[
X_{\gamma \delta} = \frac{2}{\epsilon} \sum_{\xi} \langle \psi_n | \hat{d}_{n\xi} \hat{J}_\gamma \rangle \langle \psi_n | \hat{d}_{n\xi} \hat{J}_\delta \rangle
\]
where \( \hat{d}_{n\xi} \) is given by (16) and (18).

Note that this is \( \gamma \) and \( \delta \) negative, as required by \( J^2 \) symmetry, and for a spin polarizability of the order \( 10^{-24} \) \( C^2 \) for \( \epsilon = 10^{13} \) \( \text{s}^{-1} \), and for a transition frequency \( \omega_n \) or \( 10^6 \) radians \( s^{-1} \), its order of magnitude is \( 10^{-9} \) \( C^2 \text{m}^{-3} \), which is of the order of magnitude of the molecular electric hyperpolarizability \( \gamma \). In fact, the experimentally observed electric hyperpolarizability of methane is about \( 10^{-5} \) \( C^2 \text{m}^{-3} \), which is also \( (24) \). However, the tensor differs from the customary \( \gamma \) because the latter is observable both in chiral and achiral ensembles, whereas the former is only in chiral ensembles in the absence of any \( \hat{F} \) violating electromagnetic interactions.
the 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 polarizability, hyperpolarizability, and so on) are finite both in chiral and achiral ensembles. However, if the external influence is \( \pi_i \) and the observable is \( \alpha_{ij} \), with opposite P symmetries, the effect is sustained only in chiral ensembles, and mediating P and T-negative tensors such as \( \chi_{ijk} \) 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 \( \delta_{i} \), the magnetic dipole moment induced by \( \pi_i \) is expressed as

\[
\delta_{i} = \gamma_{ij} \pi_j + \cdots \tag{30}
\]

and perturbation theory gives

\[
\gamma_{ij} = -\frac{2\pi}{\epsilon} \sum_{k} \frac{\langle \chi_{ik} \rangle}{\omega_{k}^{2}} \tag{31}
\]

This result is similar to eqn. (15) of Magne's (6) for the same magnetic contribution to the inverse Faraday effect, where the equivalent of spin polarisability is expressed, after saturation averaging, through the vector product of transition electric dipole moments.

Eqs. (30) and (31) compare directly the new effect of this paper, and the inverse Faraday effect. The latter occurs in it ensembles by principle three because \( \gamma_{ij} \) mediates the influence \( \pi_j \) and the observable \( \delta_i \), both P positive, T negative quantities. In quantum mechanics, \( \delta_i \) is also purely complex, so that \( \gamma_{ij} \) 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 (37), which is of the order of the molecular electric first hyperpolarizability. Maguire has estimated that magnetization due to the inverse Faraday effect is of the order 10⁻⁶ for a N₂/N₂ mix delivering 10⁹ W cm⁻². That of inverse magnetothermal birefringence is about 10⁻⁸, equivalent to a magnetic induction of about 10⁻⁶ 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 polarization effect introduced here appears to have many interesting consequences.

4. Discussion

The mediating tensor $\mathbf{\chi}^{(2)}$ can be amplified considerably by resonance, when the probe laser (a frequency component of broad band probe radiation) satisfies the resonance condition

$$\omega = \omega_n,$$

so that the denominators in (17) and (18) become very small. Therefore by tuning the probe to $\omega_n$ and using intense pulses of the pump, the magnitude of the induced electric dipole moment in a thin ensemble is increased by orders of magnitude. This technique combines probe resonance with the effect of the time-independent $\mathbf{P}$ 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 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 $\omega_n$. This is particularly so when attempting to amplify the minute absorptions due to electronic transitions...
related by the mutual vector dipole (25). These violate parity and allow u to u and g to g transitions which violate
approximate rule (36). In consequence, there is a very small Xij in ADAMAR ensembles due to these elementary electroweak forces,
it may be possible to annihilate the P violating Xij at a
resonance frequency of a Leptons disallowed transition in
a chiral atomic or molecular ensemble by tuning both the pump
or probe to this frequency and using the full power of the pump
or pulse trains.

We have seen in this paper that the conjugate product of
a pump laser, allowing pulses of great power, induces in
chiral ensembles a time independent electric dipole moment
through the tensor Xij, with order of magnitude and units of
electric susceptibility electric susceptibility. Therefore, it can
be used to investigate the general electric polarization
properties of chiral ensembles, using the laser instead of
electric fields. 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 E instead of a static electric field, with
availability in pulse trains of electric field strengths of
to 10^9 volts cm^-1. Stark splitting in the microwave could
therefore be observed in detail with E from a klystron laser, and
has been shown elsewhere (38-39) that E induces circular
refractive index, for example Faraday N, D, and C terms and
comparing Zeeman splitting in chiral and chiral ensembles,
d also produces forward backward interference under
appropriate conditions (32).

The effect introduced here must be distinguished
refractively from the optical Kerr effect, which elliptically illu-
scates 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
real and chiral ensembles. The Kerr effect due to 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 \( E \) applied perpendicular to the probe. In analogy the optical Kerr effect is also linear dichroism, and in contrast the effect due to \( \mathbf{E} \) introduced here is accompanied by both circular (3A) and forward backward (3B) birefringence and dichroism.

Finally in this paper, we note that the effect of \( \mathbf{E} \) on the molecular dynamics of an \( \mathbf{S} \) anisotam of perchlorofluoromethane has been computed using classical equivalents of the tensor \( \chi^{(3)} \), showing pronounced effects (35) on the orientational and rotational velocity time correlation functions, respectively Fourier transforms of the frequency dependence of dielectric loss in the Hz \( \sim \) Thz range, and of the far infra red frequency dependence in the Hz \( \sim \) 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 (15) and (22).

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