# Axial birefringence due to intense electromagnetic fields: electric and magnetic rectification

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**The Maxwell equation is solved for axial birefringence due to electric and magnetic rectification measured with a collinear unpolarized probe laser by switching an intense circularly polarized pump laser from right to left. Axial birefringence is caused by vector products of electric and magnetic conjugate components of the intense electromagnetic field. Axial birefringence can be measured with a modified Rayleigh refractometer and depends on new three- and four-rank molecular property tensors.**

It was first shown theoretically in 1982 by Wagnière and Meier[1–3] that the refractive and absorption indices of a chiral molecular ensemble are different when measured with an unpolarized probe laser parallel and antiparallel to a static magnetic field. This effect was later termed magnetochiral birefringence by Barron and Vrbancich,[4] who calculated its order of magnitude in terms of three- and four-rank molecular property tensors on which the birefringence is dependent. Other axial birefringence effects exist that are caused, for example, by a low-frequency alternating electric field in chiral ensembles and by intense circularly polarized electromagnetic fields perpendicular to a weak, unpolarized, probe laser. First-order electromagnetic axial birefringence can be treated theoretically by time averaging and by using a first-order Langevin function. In this Letter the Maxwell equation is solved for second-order axial birefringence due to electric and magnetic rectification in an intense circularly polarized pump (or power) laser parallel with the unpolarized probe laser. This effect is likely to be greater in magnitude than axial birefringence owing to a static magnetic field.[5]

Circular and axial birefringence effects caused by a pump laser, as treated here, may be of interest in the development of all-optical switch devices that rely on the nonlinear properties of inorganic and organic materials.[6–8]

In quantum perturbation theory, electric rectification is described after isotropic averaging[9] by the following product of molecular electric dipole ($\boldsymbol{\mu}$) matrix elements and field terms:

$$(\boldsymbol{\mu}\cdot\boldsymbol{\mu}'\times\boldsymbol{\mu}'')\,(\mathbf{E}_-\times\mathbf{E}_+). \tag{1}$$

The vector (or cross) product in Eq. (1) is between complex conjugates of right or left circularly polarized electric components of the intense electromagnetic field. Thus

$$\mathbf{E}_+^{(L)}\times\mathbf{E}_-^{(L)} = -\mathbf{E}_+^{(R)}\times\mathbf{E}_-^{(R)} = 2iE_0^{\,2}\mathbf{k}, \tag{2}$$

which is a product that removes the time and wave-vector dependence of the electromagnetic field and that is in general complex. The inverse Faraday effect depends after isotropic averaging[9] on the real cross product of electric conjugates,

$$i\mathbf{E}_+^{(L)}\times\mathbf{E}_-^{(L)} = -i\mathbf{E}_+^{(R)}\times\mathbf{E}_-^{(R)} = -2E_0^{\,2}\mathbf{k}. \tag{3}$$

Similarly, there are magnetic equivalents dependent on the corresponding imaginary and real vector products of magnetic field conjugates, products that again remove the phase (the exponential term) of the electromagnetic field. The imaginary product of this type is called here magnetic rectification (effect three), and the real product is called effect four. It is shown here that all four effects cause axial birefringence through new third-rank molecular property tensors in chiral ensembles.

Restricting consideration for the moment to effect one, i.e., electric rectification, it is assumed that it is mediated[1–4] by complex molecular property tensors described as

$$\mu_i = \alpha_{1ij}E_j + \alpha_{2ij}B_j + \frac{1}{3}A_{ijk}\dot{E}_{jk} + \ldots. \tag{4}$$

Here $E_j$ and $B_j$ are the electric and magnetic components of the intense pump laser, $\dot{E}_{jk}$ is the electric field gradient, $\alpha_{1ij}$ is the molecular polarizability, $\alpha_{2jk}$ is the molecular Rosenfeld tensor, and $A_{ijk}$ is a three-rank tensor[4] that mediates the induction of $\mu_i$ by $\dot{E}_{jk}$. The magnetization due to $E_j$ is written, following Ref. 4, as

$$m_i = \alpha_{2ji}^{\mathrm{conj}}E_j + \ldots, \tag{5}$$

where $\alpha_{2ij}^{\mathrm{conj}}$ is the complex conjugate of $\alpha_{2ij}$. Finally, note that the tensor $A_{kij}$ mediates the induction[4] of a molecular electric-quadrupole moment by $E_j$ of the pump laser,

$$\Theta_{ij} = A_{kij}E_k + \ldots. \tag{6}$$

The molecular polarizability tensor ($\alpha_{1ij}$) and electric–magnetic (Rosenfeld[1–4]) tensor ($\alpha_{2ij}$) are perturbed as

$$\alpha_{1ij} \rightarrow \alpha_{1ij} \pm 2\alpha_{1ijz}E_{0z}^{\,2}i \tag{7}$$

and

$$\alpha_{2ij} \rightarrow \alpha_{2ij} \pm 2\alpha_{2ijz}E_{0z}^{\,2}i \tag{8}$$

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as the polarization of the power laser is switched[10] from left to right. Similarly, the three-rank complex tensor $A_{ijk}$ is perturbed through

$$A_{ijk} \rightarrow A_{ijk} \pm 2A_{ijkl}E_{0z}{}^2 i. \qquad (9)$$

The bulk polarization, **P**, to be used in the Maxwell equation is therefore

$$\begin{aligned}\mathbf{P} = {} & N[\alpha_{1ij} \pm 2i(\alpha'_{1ijz} + i\alpha''_{1ijz})E_{0z}{}^2]E_j^{\text{probe}} \\ & + N[\alpha_{2ij} \pm 2i(\alpha'_{2ijz} + i\alpha''_{2ijz})E_{0z}{}^2]B_j^{\text{probe}} \\ & + \frac{N}{3}[A_{ijk} \pm 2i(A'_{ijkz} + iA''_{ijkz})E_{0z}{}^2)\tilde{E}_{jk}^{\text{probe}}, \qquad (10)\end{aligned}$$

where $N$ is the number of molecules per cubic meter and $E_j{}^{(p)}$ and $B_j{}^{(p)}$ are the electric and magnetic field components of the unpolarized probe laser collinear in the $z$ axis with the circularly polarized pump (or power) laser. In this expression, single and double primes denote the real and imaginary parts,[4] respectively, of the complex molecular property tensors. The bulk magnetization appearing in the Maxwell equation is

$$\mu = N[\alpha_{2ij}^{\text{conj}} \pm 2i(\alpha'^{\text{conj}}_{2ijz} + i\alpha''^{\text{conj}}_{2ijz})E_{0z}{}^2]E_j^{\text{probe}}. \qquad (11)$$

With these definitions we solve the Maxwell equation as given by Barron and Vrbancich,[4] who provide [their Eqs. (3.8a) and (3.8b)] the following equations for the real and imaginary parts of the refractive index of the ensemble in an unpolarized probe:

$$n' \doteq 1 + \frac{\mu_0}{4} c^2 N(\alpha'_{1xx} + \alpha'_{1yy} + \zeta'_{xxz} + \zeta'_{yyz}), \qquad (12)$$

$$n'' \doteq -\frac{1}{4} \mu_0 c^2 N(\alpha''_{1xx} + \alpha''_{1yy} + \zeta''_{xxz} + \zeta''_{yyz}), \qquad (13)$$

where

$$\zeta'_{\alpha\beta\gamma} = \frac{1}{c}\left[\frac{1}{3}\omega(A''_{\alpha\beta\gamma} + A''_{\beta\alpha\gamma}) + \epsilon_{\delta\gamma\alpha}\alpha'_{2\beta\delta} + \epsilon_{\delta\gamma\beta}\alpha'_{2\alpha\delta}\right], \qquad (14)$$

$$\zeta''_{\alpha\beta\gamma} = -\frac{1}{c}\left[\frac{1}{3}\omega(A'_{\alpha\beta\gamma} - A'_{\beta\alpha\gamma}) + \epsilon_{\delta\gamma\alpha}\alpha''_{2\beta\delta} - \epsilon_{\delta\gamma\beta}\alpha''_{2\alpha\delta}\right]. \qquad (15)$$

Here $\epsilon_{\delta\gamma\alpha}$ is the Levi–Civita symbol and tensor notation is used; $\mu_0$ is the permeability in vacuum and $c$ is the speed of light.

If we use an unpolarized probe laser, it measures a mean refractive index that is different for a left or right circularly polarized pump laser. This is axial birefringence due to electric rectification and, after isotropic averaging, is given from relations (12) and (14) by

$$\langle n'_{\uparrow\uparrow\mathrm{R}} - n'_{\uparrow\uparrow\mathrm{L}}\rangle_1 = 2\mu_0 c N E_{0z}{}^2\left[\langle\alpha''_{2xyz}\rangle + \langle\alpha''_{2yxz}\rangle - \frac{\omega}{3}(\langle A'_{xxzz}\rangle + \langle A'_{yyzz}\rangle)\right] + \mathrm{o}\,(\text{higher}).... \qquad (16)$$

The scalar components that survive ensemble averaging are obtained from theorems in the literature.[4,11] These are odd-parity tensor components, which are supported in consequence only by chiral ensembles.

Axial birefringence due to electric rectification is mediated by these molecular property tensors, which are in general complex and frequency dependent. Equation (16) is, essentially, a new type of spectrum that depends on the square of the electric field intensity ($E_0$) of the pump laser through these new frequency-dependent antisymmetric scalar components of three- and four-rank property tensors that mediate the effect of the field conjugate product $\mathbf{E}_+ \times \mathbf{E}_-$ on the Rosenfeld tensor $\alpha_{2ij}$ and the electric-dipole–electric-quadrupole tensor $A_{ijk}$.

Similarly, there is axial birefringence due to the complex field product $i\mathbf{E}_+ \times \mathbf{E}_-$ that is responsible[9] for the inverse Faraday effect of nonlinear optics. It is given by

$$\langle n'_{\uparrow\uparrow R} - n'_{\uparrow\uparrow L}\rangle_2 = 2\mu_0 c N E_{0z}{}^2\left[\langle\alpha'_{2xyz}\rangle + \langle\alpha'_{2yxz}\rangle - \frac{\omega}{3}(\langle A''_{xxzz}\rangle + \langle A''_{yyzz}\rangle)\right] + \ldots, \qquad (17)$$

i.e., by the real parts, as opposed to the imaginary parts [Eq. (16)], of the new tensor $\alpha_{2ij}$ and vice versa for $A_{ijk}$. This is called effect two. Effects three and four are found in an entirely analogous way and are due, respectively, to the magnetic field conjugate products $\mathbf{B}_+ \times \mathbf{B}_-$ and $i\mathbf{B}_+ \times \mathbf{B}_-$. In general, all four of these second-order (nonlinear) effects are present simultaneously.

Symmetry arguments[12] for these effects have been given, where they were classified as spin-chiral effects. In analogy with magnetochiral dichroism,[4] the axial birefringence proposed here can be measured in principle by using a modified Rayleigh refractometer.[4] Left and right circularly polarized power laser radiation from, for example, a terawatt laser pulse train, is sent simultaneously down the two arms of the refractometer that is filled with a chiral material. The birefringence is measured with a weak, unpolarized, tunable probe laser at a fixed frequency of the pump laser, using standard, time-resolved, phase-sensitive detection techniques. The radiation from the intense pump laser is filtered and prevented from reaching the detector. The same filter permits the radiation from the probe laser at different frequencies to pass through, thus the axial birefringence can be isolated and measured directly.

All four axial birefringence effects are second order, either in $E_{0z}{}^2$ or $B_{0z}{}^2$; to estimate the order of magnitude of effect one, for example, we need to use the appropriate Kielich function[13,14] to estimate the ensemble thermodynamic averages,

$$\langle\alpha''_{2xyz}\rangle, \langle\alpha''_{2yxz}\rangle, \langle A'_{xxzz}\rangle, \langle A'_{yyzz}\rangle,$$

and so on. The Kielich function has an upper bound of unity (saturation), which can be achieved[14] by a powerful pulse of a focused giant ruby pump laser. In this event, an axial birefringence of the order 0.1 is achievable in theory, which is easily within range of the Rayleigh refractometer. This is several orders of magnitude larger than magnetochiral birefringence.

An order-of-magnitude estimate of the effect can be made by writing Eq. (17), for example, as

$$\langle n'_{\uparrow\uparrow R} - n'_{\uparrow\uparrow L}\rangle_2 \doteq 2\mu_0 c N E_{0z}^{\,2} \alpha'_{2xy} L_2 + \ldots, \tag{18}$$

where $L_2$ is the second-order Kielich function[15] that mediates the orienting effect of the pump laser, which sets up a potential energy of the form

$$-\alpha_{1ij} E_{0i} E_{0j}$$

with the molecular polarizability. The torque set up by this effect is computer simulated in Ref. 16. The Kielich function goes to unity as $E_0 \to \infty$. We set this conservatively at 0.01. The other quantities in Eq. (17) are $\mu_0 = 4\pi \times 10^{-7}$ J sec$^2$ C$^{-2}$ m$^{-1}$, $c = 3 \times 10^8$ m sec$^{-1}$, and $N = 6 \times 10^{26}$ m$^{-3}$; for $\alpha'_{2xy}$ we use[4] a conservative order of magnitude of $10^{-36}$ $A^2$ J$^{-1}$ m$^3$ sec. This leaves the electric-field strength of the pump laser. Hutchinson[17] estimates that this can reach approximately $10^9$ V/m in a small, commercially available, Q-switched and focused Nd:YAG laser. For $L_2 = 0.01$, the birefringence from relation (18) is

$$\langle n'_{\uparrow\uparrow R} - n'_{\uparrow\uparrow L}\rangle \doteq 144\pi \times 10^{-11} E_{0z}^{\,2} + \ldots, \tag{19}$$

and for a conservative value of $E_{0z}$ of 1000 V/m this is approximately 0.01. Clearly, in a focused and Q-switched Nd:YAG laser, the effect can be orders of magnitude larger, and the Kielich function $L_2$ can be saturated, i.e., reach 1.0.

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