AXIAL BIREFRINGENCE DUE TO ELECTROMAGNETIC FIELDS: SPIN CHIRAL EFFECTS

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The Maxwell equation is solved for axial birefringence caused by a circularly polarised power laser parallel and anti-parallel to a weak unpolarised probe laser. This class of electromagnetically induced axial birefringence is called "spin chiral", mediated by new third rank molecular property tensors. The spin chiral class of axial birefringence is due to vector products of electric and magnetic conjugate components of the intense, circularly polarised, electromagnetic field.

1. Introduction

The first axial birefringence phenomenon was analysed by Wagnière and Meier [1–3] in 1982, and is caused by a static magnetic field parallel or anti-parallel with an unpolarised probe electromagnetic field in the Z axis of the laboratory frame. It was called "magneto-chiral" birefringence by Barron and Vrbancich [4] and is mediated by a parity-odd third rank molecular property tensor, which is non-zero only in a chiral molecular ensemble [5–8]. Its dynamical basis was analysed by Evans [9] using group theoretical statistical mechanics [10–12]. This symmetry analysis anticipated the presence of an analogous axial birefringence with the static magnetic field replaced by a circularly polarised power laser, an effect which was called "spin chiral dichroism and birefringence" [9]. Kostov and Zheludev [13] have recently made the interesting discovery, related to axial birefringence, that there is a rotation of the polarisation plane of a light beam normally reflected from a cubic crystal, a composite superlattice of GaAs. This technique [13] may be potentially useful for the observation of the non-linear effect introduced here.

In this Letter the Maxwell equation is solved for a new type of axial birefringence, which is shown to be mediated by new third rank, frequency dependent, complex, molecular property tensors. Two types of spin chiral birefringence are discussed, depending respectively on the real and imaginary vector products $\boldsymbol{E}\times\boldsymbol{H}$ and $\mathrm{i}\boldsymbol{E}\times\boldsymbol{H}$ of complex conjugate electric and magnetic components of the intense, circularly polarised, electromagnetic power field.

2. The conjugate field products

The conjugate field products responsible for spin chiral axial birefringence are those described by Wagnière [14] in the contexts of parity violation and inverse magneto-chiral birefringence. Quantum perturbation theory [14] shows that the latter is magnetization in chiral ensembles due to the real conjugate product

$$\begin{aligned}&\boldsymbol{E}_+^{(\mathrm{L})}\times\boldsymbol{H}_-^{(\mathrm{L})}+\boldsymbol{E}_-^{(\mathrm{L})}\times\boldsymbol{H}_+^{(\mathrm{L})}\\&=\boldsymbol{E}_+^{(\mathrm{R})}\times\boldsymbol{H}_-^{(\mathrm{R})}+\boldsymbol{E}_-^{(\mathrm{R})}\times\boldsymbol{H}_+^{(\mathrm{R})}\\&=4E_0H_0\boldsymbol{k}\end{aligned}\tag{1}$$

of electric and magnetic components of an intense, circularly polarised electromagnetic field. The imaginary conjugate product

$$\begin{aligned}&\mathrm{i}(\boldsymbol{E}_+^{(\mathrm{L})}\times\boldsymbol{H}_-^{(\mathrm{L})}+\boldsymbol{E}_-^{(\mathrm{L})}\times\boldsymbol{H}_+^{(\mathrm{L})})\\&=\mathrm{i}(\boldsymbol{E}_+^{(\mathrm{R})}\times\boldsymbol{H}_-^{(\mathrm{R})}+\boldsymbol{E}_-^{(\mathrm{R})}\times\boldsymbol{H}_+^{(\mathrm{R})})\\&=4\mathrm{i}E_0H_0\boldsymbol{k}\end{aligned}\tag{2}$$

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induces an electric polarisation [14] which is shown here also to result in axial birefringence. The latter is observable in both cases by measuring with a weak, unpolarised probe laser the refractive index of a molecular ensemble, the probe being parallel and anti-parallel with a circularly polarised power laser. This is possible with a modified Rayleigh refractometer.

3. The molecular property tensors

It is assumed that the molecular polarisability tensor,

$$\alpha_{1ij} \equiv \alpha'_{1ij} + i\alpha''_{1ij} \tag{3}$$

and Rosenfeld (natural optical activity mediating) tensor [15],

$$\alpha_{2ij} \equiv \alpha'_{2ij} + i\alpha''_{2ij} \tag{4}$$

are modified as follows by the real and imaginary conjugate products of section (1),

$$\alpha^{\{1\}}_{1ij} = \alpha_{1ij} \pm 4(\alpha'_{3ijZ} + i\alpha''_{3ijZ})\ (E_0H_0)_Z\,, \tag{5}$$

$$\alpha^{\{1\}}_{2ij} = \alpha_{2ij} \pm 4(\alpha'_{4ijZ} + i\alpha''_{4ijZ})\ (E_0H_0)_Z \tag{6}$$

and

$$\alpha^{\{2\}}_{1ij} = \alpha_{1ij} \pm 4i(\alpha'_{3ijZ} + i\alpha''_{3ijZ})\ (E_0H_0)_Z\,, \tag{7}$$

$$\alpha^{\{2\}}_{2ij} = \alpha_{2ij} \pm 4i(\alpha'_{4ijZ} + i\alpha''_{4ijZ})\ (E_0H_0)_Z\,. \tag{8}$$

The plus signs in these expressions indicate a circularly polarised laser parallel with the weak, unpolarised probe laser, and the minus signs anti-parallel. The third rank tensors [4] mediate the effects of the real and imaginary conjugate products on the polarisability and Rosenfeld tensors. In consequence, the polarisation, $\boldsymbol{P}$, of the Maxwell equation is

$$P_i = N(\alpha^{\{1\}}_{ij} E^{\text{probe}}_j + \alpha^{\{1\}}_{2ij} B^{\text{probe}}_j) \tag{9}$$

for the real conjugate product (responsible [14] for inverse magneto-chiral birefringence) and

$$P_i = N(\alpha^{\{2\}}_{ij} E^{\text{probe}}_j + \alpha^{\{2\}}_{2ij} B^{\text{probe}}_j) \tag{10}$$

for the imaginary [14]. Here E^{probe}_j and B^{probe}_j are the electric and magnetic field components of the probe laser. The solution of the Maxwell equation for the axial birefringence in both cases proceeds as follows.

4. Solving the Maxwell equation

We use the standard Maxwell equation

$$\frac{1}{\mu_0}\nabla \times \boldsymbol{B}^{\text{probe}} = \epsilon_0 \frac{\partial \boldsymbol{E}^{\text{probe}}}{\partial t} + \frac{\partial \boldsymbol{p}}{\partial t}\,, \tag{11}$$

where μ_0 and ϵ_0 are respectively the permeability and permittivity in vacuo, and solve it [16–18] for the mean, real, refractive index of the molecular ensemble as measured by the unpolarised probe in the Y axis of the laboratory frame. For the real conjugate product this is

$$n'_{\text{av}} = \mu_0 c\epsilon_0 \frac{E^{\text{probe}}_0}{B^{\text{probe}}_0} + N\Bigg(\mu_0 c \frac{E^{\text{probe}}_0}{B^{\text{probe}}_0}\,[\alpha'_{1YY} \pm 4\alpha'_{3YYZ}(E_0H_0)_Z] - \mu_0 c[\alpha'_{2YX} \pm 4\alpha'_{4YXZ}(E_0H_0)_Z]\Bigg) \tag{12}$$

and for the imaginary conjugate product

$$n'_{\text{av}} = \mu_0 c\epsilon_0 \frac{E^{\text{probe}}_0}{B^{\text{probe}}_0} + N\Bigg(\mu_0 c \frac{E^{\text{probe}}_0}{B^{\text{probe}}_0}\,[\alpha'_{1YY} \mp 4\alpha''_{3YYZ}(E_0H_0)_Z] - \mu_0 c[\alpha'_{2YX} \mp 4\alpha''_{4YXZ}(E_0H_0)_Z]\Bigg). \tag{13}$$

In each case it is seen that this mean refractive index is different parallel and anti-parallel with the circularly (left or right) polarised power laser. This difference is termed spin chiral axial birefringence and after ensemble averaging [4] is given as follows for the real and imaginary field conjugate products respectively,

$$\langle n'_{\text{av}}(\uparrow\uparrow) - n'_{\text{av}}(\uparrow\downarrow)\rangle_{\text{R}} = -8N\mu_0 c(E_0H_0)_Z \langle \alpha'_{4YXZ}\rangle \tag{14}$$

and

$$\langle n'_{av}(\uparrow\uparrow) - n'_{av}(\uparrow\downarrow)\rangle_I = 8N\mu_0 c(E_0H_0)_Z \langle \alpha''_{4YXZ}\rangle , \quad (15)$$

i.e. through the totally anti-symmetric real and imaginary scalar components of the third rank mediating complex tensor α_{4ijk}.

5. Considerations of parity

The tensor α_{4ijk} multiplies a conjugate field product which is parity-odd. It must therefore be parity-even to match the odd parity of the Rosenfeld tensor on the left hand side of eq. (6). The parity-even third rank tensor α_{4ijk} is supported in consequence both by chiral and achiral molecular ensembles, and the spin chiral class of axial birefringence should be observable in both types of ensemble. This contrasts with magneto-chiral birefringence, which can be supported by chiral ensembles only [1–4]. Other types of axial birefringence also vanish in achiral ensembles, such as that due to a low frequency alternating electric field [16], and electromagnetically induced axial birefringence due to electric and magnetic rectification [18]. These three effects all depend on odd-parity third rank molecular property tensors, in contrast with the spin chiral class considered here.

6. Experimental observation and approximate order of magnitude

The spin chiral type of axial birefringence is measured by a weak, unpolarised, probe laser, or ordinary light beam, parallel and anti-parallel with a circularly polarised power laser, such as a giant ruby laser [19]. The frequency dependence of spin chiral axial birefringence is a specific source of information on the molecular property tensor α_{4ijk}, and is a new type of spectroscopy.

One convenient way of measurement is by sending a pulse train from the power laser, at one fixed frequency, in opposite directions down the two arms of a Rayleigh refractometer, as first suggested [4] for magneto-chiral birefringence. This could be achieved accurately with contemporary beam splitters and mirrors. After passage through the refractometer, the ruby laser radiation is filtered out completely before reaching the detector system. The same filter, which is opaque at the ruby laser frequency, is transparent at most frequencies of the probe. The latter reaches a phase sensitive, time resolved, detector system, which directly measures changes in the refractive index of a molecular material in the Rayleigh refractometer due to a pulse train of the giant ruby laser. The magnitude of the effect should be proportional to the product of intensities $(E_0H_0)_Z$ of the giant ruby laser, i.e. is a mixed second-order effect. It is well known [19] that a giant ruby laser pulse can saturate the Langevin function, and this is expected to produce [17] axial birefringence several orders of magnitude greater than magneto-chiral birefringence. The latter is itself within range of the Rayleigh refractometer [4], so that the spin chiral class should be easily observable in an ensemble such as water. The spectral information carried by the unpolarised, broad-band, probe radiation can be analysed conveniently by contemporary Fourier transform methods [20], providing the specific spectral response of the absorption index (complex part of the refractive index).

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