Optics Communications 346 (2015) 80–87

Contents lists available at ScienceDirect

Optics Communications

journal homepage: www.elsevier.com/locate/optcom

Measurement of quadratic electrogyration effect in castor oil

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ARTICLE INFO

Article history:
Received 31 August 2014
Received in revised form
23 January 2015
Accepted 7 February 2015
Available online 9 February 2015

Keywords:
Electrogyration effect
Dichroism
Optical activity
Linear birefringence
Electro-optic effect
Castor oil

ABSTRACT

This work presents a detailed analysis of electrogyration measurement in liquids with the usage of an optical polarimetric technique. Theoretical analysis of the optical response to an applied electric field is illustrated by experimental data for castor oil which exhibits natural optical activity, quadratic electro-optic effect and quadratic electrogyration effect. Moreover, the experimental data show that interaction of the oil with a pair of flat electrodes induces a significant dichroism and natural linear birefringence. The combination of these effects occurring at the same time complicates the procedure of measurements. It has been found that a single measurement is insufficient to separate the contribution of the electrogyration effect, but it is possible on the basis of several measurements performed with various orientations of the polarizer and the analyser. The obtained average values of the quadratic electrogyration coefficient β_{13} in castor oil at room temperature are from -0.92×10^{-22} to -1.44×10^{-22} $m^2 V^{-2}$ depending on the origin of the oil. Although this study is focused on measurements in castor oil, the presented analysis is much more general.

1. Introduction

Optical activity induced by an applied electric field, known as electrogyration effect, was the subject of many papers published for over 50 years. The results known from the literature, however, focus on the effect in solid [1–6] and liquid crystals [7–10], but it is highly difficult to find any published experimental data concerning the liquids which are not liquid crystals. Moreover, we have found that the measurement methods described previously for solid and liquid crystals may not be suitable for some liquids. Among the liquids widely used in the industry, some vegetable oils, e.g. castor oil, are optically active and exhibit a particularly complicated combination of various optical effects occurring simultaneously. Castor oil has a lot of applications in medicine, cosmetics, food industry, as an addition to biodiesel, as lubricant, brake fluid, dielectric fluid and many more. However, low oxidative and temperature stability limits its shelf life and widespread use. The demand to control the purity of vegetable oils also resulted in seeking for the optical constants which exhibit high sensitivity to changes in the composition of the oil. Recent studies have shown that a particularly promising constants are Verdet constant that describes the strength of magneto optical effect and may be used in authentication technique for olive oil and probably also other vegetable oils [11] and Kerr constant describing quadratic electrooptic effect which clearly changes under the influence of aging processes in castor oil [12]. Our initial observations indicate that the coefficients of quadratic electrogyration effect may be another optical constants suitable for the oils quality control.

According to our best knowledge, no general method for the measurements of electrogyration in liquids has been reported so far. The aim of this paper is to show with the example of castor oil that quadratic electrogyration effect is measurable in a liquid which is not a liquid crystal and it is possible to separate this effect from the much stronger electro-optic effect manifesting both directly and in some complex mechanisms of light modulation involving a dichroism, linear birefringence and natural optical activity. At the present stage of research, we focus on developing a number of physical, mathematical and technical issues in the measurement. Our main long-term motivation is, however, a need to build a system for reliable and continuous assessment of the quality of castor oil based on measurements of a number of optical constants without any chromatographic and chemical analyzes. Moreover, it seems important to check whether the quadratic electrogyration is so strong that it can significantly disturb previous measurements of the Kerr constant in vegetable oils. An additional motivation for our work is a need to choose an immersion liquid that does not exhibit the electrogyration effect, which is important in precise measurements of electrogyration in solid crystals. Our results show that the methyl silicone oil OM50, which was taken in this study as a control liquid, may be a suitable

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http://dx.doi.org/10.1016/j.optcom.2015.02.013
0030-4018/

immersion liquid.

The correct mathematical description of the optical properties of a given liquid in an applied electric field requires knowledge of the Curie group describing the internal symmetry of the liquid. In the case of optically active liquids the possible groups are $\infty\infty$, $\infty 2$ and ∞. The castor oil in a sufficiently large sample and in the absence of any external fields can be considered as an isotropic non-dichroic liquid of $\infty\infty$ symmetry. However, the same oil placed between two flat metal electrodes spaced a few millimeters begins to exhibit some new effects such as dichroism and linear birefringence, which indicates the transition to $\infty 2$ or ∞ symmetry. In our previous work [13] we presented results showing that these effects may by induced in the oil by a pair of stainless steel plane-parallel plates, even without any applied electric field. Our present measurements with the use of an electric field allow detection of minimal disturbance of the natural $\infty\infty$ symmetry of castor oil and distinguish clearly between $\infty 2$ and ∞ symmetries.

2. Theoretical analysis

2.1. Impermeability tensor

The electric displacement **D** in a homogeneous, non-absorbing, non-magnetic and optically active medium caused by an electric field $\mathcal{E}$ can be written as [14–16]

$$\mathbf{D} = \varepsilon_0\{[\varepsilon_r]\mathcal{E} + \mathrm{i}\mathbf{G}\times\mathcal{E}\} = \varepsilon_0\{[\varepsilon_r] + \mathrm{i}[G]\}\mathcal{E} = \varepsilon_0[K]\mathcal{E}, \quad (1)$$

where ε_0 is the vacuum permittivity, i denotes the imaginary unit, **G** is the real gyration vector, and $[K]$ is the relative permittivity of the medium represented by complex Hermitian tensor. If $\mathcal{E}$ in Eq. (1) refers to the electric field of the light wave, the real symmetric part $[\varepsilon_r]$ of $[K]$ represents the linear birefringence, and the antisymmetric imaginary part $[G]$ represents the optical activity.

In the case of absorbing media the symmetry $K_{ij} = K_{ji}^*$ (where * indicates complex conjugate) is not exactly fulfilled [15,17]. Although castor oil exhibits significant absorption of light, but the corresponding asymmetry of the $[K]$ tensor is very small and will be neglected in our further derivations.

The vector product $\mathbf{G}\times\mathcal{E}$ in Eq. (1) can always be represented as $[G]\mathcal{E}$ product, where $[G]$ is a real antisymmetric tensor composed of the components of the **G** vector:

$$[G] = \begin{bmatrix} 0 & -G_3 & G_2 \\ G_3 & 0 & -G_1 \\ -G_2 & G_1 & 0 \end{bmatrix}. \quad (2)$$

The gyration vector **G** for a given wave propagation vector **s** can be calculated as

$$\mathbf{G} = [g]\mathbf{s}, \quad (3)$$

where $[g]$ is a second rank gyration axial tensor described by a real 3×3 matrix. All the possible symmetries $\infty\infty$, $\infty 2$ and ∞ of the liquids that exhibit natural optical activity also enable the activity induced by an applied DC or low-frequency electric field **E** and the total optical activity can be expressed by the following power series:

$$g_{ij} = g_{ij}^{(0)} + \gamma_{ijk}E_k + \beta_{ijkl}E_kE_l + \cdots, \quad (4)$$

where $g_{ij}^{(0)}$ are the components of the gyration tensor describing a natural optical activity, and γ_{ijk} and β_{ijkl} are the components of the linear and quadratic electrogyration tensors, respectively.

The coefficients of electro-optic effects may be defined through changes in the real part of the permittivity tensor in response to an applied electric field **E**, but more usual approach involves impermeability tensor defined as $[B] = [K]^{-1}$. The real symmetric part of the $[B]$ tensor at an optical frequency

$$\mathrm{Re}[B_{ij}] = \mathrm{Re}[B_{ij}^{(0)}] + r_{ijk}E_k + q_{ijkl}E_kE_l + \cdots, \quad (5)$$

where $\mathrm{Re}[B_{ij}^{(0)}]$ are the components of the tensor related to the natural linear birefringence, and r_{ijk} and q_{ijkl} are the components of the linear and quadratic electro-optic tensors, respectively. The imaginary antisymmetric part of $[B]$ may be found from the relationship [18] which results from the definition $[B] = [K]^{-1}$:

$$\mathrm{Im}[B] = -(\mathrm{Re}[B])[G](\mathrm{Re}[B]). \quad (6)$$

Table 1
The form of the gyration tensor $[g_{ij}]$ for selected $\infty 2$ and ∞ Curie groups in the presence of an electric field [17]. In the both groups $\beta_{66} = (1/2)(\beta_{11} - \beta_{12})$.

$g_{ij} = g_{ij}^{(0)} + \gamma_{ijk}E_k + \beta_{ijkl}E_kE_l + \cdots$		
Natural optical activity $g_{ij}^{(0)}$	Linear electrogyration γ_{ijk}	Quadratic electrogyration β_{ijkl}
$\infty 2$ Curie group		
$\begin{bmatrix} g_{11}^{(0)} & 0 & 0 \\ 0 & g_{11}^{(0)} & 0 \\ 0 & 0 & g_{33}^{(0)} \end{bmatrix}$	$\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ \gamma_{41} & 0 & 0 \\ 0 & -\gamma_{41} & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{bmatrix} \beta_{11} & \beta_{12} & \beta_{13} & 0 & 0 & 0 \\ \beta_{12} & \beta_{11} & \beta_{13} & 0 & 0 & 0 \\ \beta_{31} & \beta_{31} & \beta_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & \beta_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & \beta_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & \beta_{66} \end{bmatrix}$
∞ Curie group		
$\begin{bmatrix} g_{11}^{(0)} & 0 & 0 \\ 0 & g_{11}^{(0)} & 0 \\ 0 & 0 & g_{33}^{(0)} \end{bmatrix}$	$\begin{bmatrix} 0 & 0 & \gamma_{13} \\ 0 & 0 & \gamma_{13} \\ 0 & 0 & \gamma_{33} \\ \gamma_{41} & \gamma_{51} & 0 \\ \gamma_{51} & -\gamma_{41} & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{bmatrix} \beta_{11} & \beta_{12} & \beta_{13} & 0 & 0 & \beta_{16} \\ \beta_{12} & \beta_{11} & \beta_{13} & 0 & 0 & -\beta_{16} \\ \beta_{31} & \beta_{31} & \beta_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & \beta_{44} & \beta_{45} & 0 \\ 0 & 0 & 0 & -\beta_{45} & \beta_{44} & 0 \\ -\beta_{16} & \beta_{16} & 0 & 0 & 0 & \beta_{66} \end{bmatrix}$

The $\infty\infty$, $\infty 2$ and ∞ Curie groups contain only rotational symmetry elements. Since the rotation around any axis does not result in a change in handedness of the Cartesian coordinate system, the forms of pseudotensors $g_{ij}^{(0)}$, γ_{ijk}, β_{ijkl} of the internal symmetries $\varepsilon[V^2]$, $(\varepsilon[V^2])[V]$, and $(\varepsilon[V^2])[V^2]$ (see Table 1) are analogous to the forms of tensors $\mathrm{Re}[B_{ij}^{(0)}]$, r_{ijk}, q_{ijkl} with the internal symmetries $[V^2]$, $[V^2][V]$ and $[V^2][V^2]$ (see Table 2), respectively.

A pair of parallel flat electrodes immersed in castor oil induce a dichroism and linear birefringence, and the optical axis that appears in the oil is perpendicular to the plane of the electrodes. Our previous experiment showed that the effect might be observed even in the absence of any voltage applied to the electrodes [13]. The latter measurements showed that dichroism and linear birefringence additionally increase after switching on the electric field. Thus, the formulas (1)–(6) and Tables 1 and 2 are fulfilled in such a coordinate system in which the Z-axis is associated with the applied electric field $\mathbf{E} = [0, 0, E]$, while the X and Y axes can be chosen freely. As the XYZ coordinates are not convenient for calculations of the light transmission through the sample we introduced another coordinate system $X'Y'Z'$ in which the direction of the light beam $\mathbf{s}\|Z'$ and the direction of the field $\mathbf{E}\|X'$. The $[B']$ tensor written for the $\infty 2$ Curie group with the accuracy to quadratic terms has the following form in the $X'Y'Z'$ coordinates: