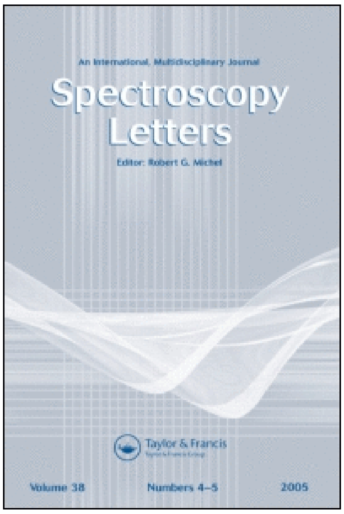

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Optical Calculi

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OPTICAL CALCULI

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Optical calculi for anisotropic media are reviewed and extended for use in phasemodulation spectroscopy.

If one needs a calculus for treatment of the interaction between light and optical systems different approaches may be used depending on the representation used for the light wave, i.e. either as Stokes or Maxwell vector[1,2]. The two calculi are known as the Mueller and Jones algebra respectively, honouring the two men who some 30 years ago developed the formalisms.

Depending on the aim of an investigation one may choose either of the two calculi since different advantages are featured by them. The choise of the spectroscopist has often been the Mueller calculus, as it can handle problems involving depolarizations and furthermore works on a light representation which, although fourdimensional, contains an element proportional to the light intensity measurable with a photomultiplier. Contrary the choise of theorists is normally the Jones calculus, as it works on a two-dimensional basis and depends on the validity of the electromagnetic theory.

For obvious reasons it is worthwhile to investigate the relations between the Mueller calculus, based on a phenomenological approach, and the Jones calculus. This kind of work has been carried out earlier by Parke[3], the result, however, is partly incorrect and is furthermore written with the Stokes' parameters arranged differently compared with the work of Schmieder[4], in which also the relation between a Jones and a Mueller matrix is found, so that given an arbitrary Jones matrix the Mueller ditto may be evaluated, but not vice versa.

It is the merit of Jones to have shown how to parametrizise with respect to the phenomenological quantities linear dichroism (LD), linear birefringence (LB), circular dichroism (CD), and circular birefringence (CB)[5] a matrix representing an optical device showing both anisotropy and optical activity. The formalism holds for light passing normally through a homogenous media with plane and parallel surfaces perpendicular on a principal axis in the index ellipsoid of the medium. The result, however, is very complex and therefore it is desirable to find a way of factorizing the matrix; this may, as indicated by Gō[6] and Troxell and Scheraga[7], be achieved by transforming the matrix from Jones into Mueller form. It is the aim of this paper to investigate this transformation process; further developments will be found elsewhere[8].

JONES' FORMALISM

The matrix representing a medium with pathlength z, given in a laboratory-fixed coordinate system with the direction of light propagation as the z-axis and the origin placed on the entrance surface, is formulated as:

$$\underline{M} = \begin{pmatrix} m_1 & m_4 \\ m_3 & m_2 \end{pmatrix} \qquad (1)$$

and $\underline{M}$ is furthermore related to a point-matrix $\underline{N}$ through the equation

$$\underline{M} = e^{\underline{N}z}$$

Analyzing the eigenvalue problem of the $\underline{M}$ and $\underline{N}$ matrices respectively, it is possible to find relations between the two sets of matrix elements e.g.

$$m_1 = f(n_1, n_2, n_3, n_4, z),$$

the total expression may be found elsewhere[5,8].

The point-matrix may now be parametrizised, and as the matrix contains four complex elements it is necessary for the purpose to use eight phenemenological parameters; these may be found in Jones' original papers[5]. Thus it turns out that the parametrizised $\underline{N}$-matrix has the following form:

$$\underline{N} = \begin{pmatrix} (-\kappa+p_o-i\eta+ig_o) & (-\omega+p_{45}-i\delta+ig_{45}) \\ (\omega+p_{45}+i\delta+ig_{45}) & (-\kappa-p_o-i\eta-ig_o) \end{pmatrix} \quad (2)$$

Before we proceed to the transformation from Jones into Mueller formalism, it is desirable to associate the eight Jones parameters with two-letter mnemonics since we in this field has a severe notational problem and no established usage.

$2\kappa z$	$2p_o z$	$2g_o z$	$2\delta z$	$-2\omega z$	$2p_{45}z$	$2g_{45}z$
Abs	LD	LB	CD	CB	LD'	LB'

MUELLER FORMALISM

We may now transform the Jones matrix ($\underline{M}$), representing under restrictions given in the introduction, the general anisotropic and optically active medium into a four-dimensional Mueller matrix. For that purpose we need the relation between the two sets of matrices, which may be obtained either as done by Parke[3] or by Schmeider[4]. To correct errors in the literature and to write the conversion consistently, we establish below the transformation of the matrix given by equation (1) to give twice the general Mueller matrix: (* □ complex conjugation).

$$\begin{pmatrix} m_1m_1^*+m_4m_4^*+m_3m_3^*+m_2m_2^* & m_4m_1^*+m_1m_4^*+m_2m_3^*+m_3m_2^* & -i(m_1m_4^*-m_4m_1^*+m_3m_2^*-m_2m_3^*) & m_1m_1^*-m_4m_4^*+m_3m_3^*-m_2m_2^* \\ m_3m_1^*+m_2m_4^*+m_1m_3^*+m_4m_2^* & m_2m_1^*+m_3m_4^*+m_4m_3^*+m_1m_2^* & -i(m_3m_4^*-m_2m_1^*+m_1m_2^*-m_4m_3^*) & m_3m_1^*-m_2m_4^*+m_1m_3^*-m_4m_2^* \\ -i(m_3m_1^*+m_2m_4^*-m_1m_3^*-m_4m_2^*) & -i(m_2m_1^*+m_3m_4^*-m_4m_3^*-m_1m_2^*) & m_1m_2^*-m_4m_3^*-m_3m_4^*+m_2m_1^* & -i(m_3m_1^*-m_2m_4^*-m_1m_3^*+m_4m_2^*) \\ m_1m_1^*+m_4m_4^*-m_3m_3^*-m_2m_2^* & m_4m_1^*+m_1m_4^*-m_2m_3^*-m_3m_2^* & -i(m_1m_4^*-m_4m_1^*-m_3m_2^*+m_2m_3^*) & m_1m_1^*-m_4m_4^*-m_3m_3^*+m_2m_2^* \end{pmatrix}$$

(3)

We may now insert the phenomenological mnemonics in Jones' point-matrix (2), calculate Jones' $\underline{M}$ matrix, and convert to Mueller formalism according to the above given expression.

Abbreviations in the matrices given below are as follows:

$a = LD^2 + CD^2 + LD'^2 - LB^2 - CB^2 - LB'^2$

$b = 2LD\cdot LB + 2CD\cdot CB + 2LD'\cdot LB'$

$$
\begin{pmatrix}
\begin{array}{l} \frac{A}{2}(ch^2B-s^2C)+ \\ \frac{1}{2}(ch^2B-c^2C)\cdot \\ (LD^2+LB^2+CD^2+ \\ \;CB^2+LD'^2+LB'^2) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LD\cdot CB-LB\cdot CD)+ \\ sh2B(B\cdot LD'+C\cdot LB') \\ +s2C(C\cdot LD'-B\cdot LB') \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LB\cdot LD'-LB'\cdot LD)+ \\ sh2B(C\cdot CB+B\cdot CD)+ \\ s2C(C\cdot CD-B\cdot CB) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (CD\cdot LB'-CB\cdot LD')+ \\ sh2B(B\cdot LD+C\cdot LB)+ \\ s2C(C\cdot LD-B\cdot LB) \end{array} \\
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LB\cdot CD-LD\cdot CB)+ \\ sh2B(B\cdot LD'+C\cdot LB') \\ +s2C(C\cdot LD'-B\cdot LB') \end{array} &
\begin{array}{l} \frac{A}{2}(ch^2B-s^2C)+ \\ \frac{1}{2}(ch^2B-c^2C)\cdot \\ (-LB^2-CD^2-LD^2- \\ CB^2+LD'^2+LB'^2) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LB'\cdot CB+CD\cdot LD')+ \\ sh2B(B\cdot LB-C\cdot LD)+ \\ s2C(B\cdot LD+C\cdot LB) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LD\cdot LD'+LB\cdot LB')+ \\ sh2B(C\cdot CD-B\cdot CB)+ \\ s2C(-C\cdot CB-B\cdot CD) \end{array} \\
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LB'\cdot LD-LB\cdot LD')+ \\ sh2B(C\cdot CB+B\cdot CD)+ \\ s2C(C\cdot CD-B\cdot CB) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LB'\cdot CB+CD\cdot LD')+ \\ sh2B(C\cdot LD-B\cdot LB)+ \\ s2C(-B\cdot LD-C\cdot LB) \end{array} &
\begin{array}{l} \frac{A}{2}(ch^2B-s^2C)+ \\ \frac{1}{2}(ch^2B-c^2C)\cdot \\ (-LD^2-LB^2+CD^2+ \\ CB^2-LD'^2-LB'^2) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LD\cdot CD+LB\cdot CB)+ \\ sh2B(B\cdot LB'-C\cdot LD')+ \\ s2C(B\cdot LD'+C\cdot LB') \end{array} \\
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (CB\cdot LD'-CD\cdot LB')+ \\ sh2B(B\cdot LD+C\cdot LB)+ \\ s2C(C\cdot LD-B\cdot LB) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LD\cdot LD'+LB\cdot LB')+ \\ sh2B(B\cdot CB-C\cdot CD)+ \\ s2C(C\cdot CB+B\cdot CD) \end{array} &
\begin{array}{l} (ch^2B-c^2C)\cdot \\ (LD\cdot CD+LB\cdot CB)+ \\ sh2B(C\cdot LD'-B\cdot LB')+ \\ s2C(-B\cdot LD'-C\cdot LB') \end{array} &
\begin{array}{l} \frac{A}{2}(ch^2B-s^2C)+ \\ \frac{1}{2}(ch^2B-c^2C)\cdot \\ (LD^2+LB^2-CB^2- \\ \;LD'^2-CD^2-LB'^2) \end{array}
\end{pmatrix}
\tag{4}
$$

$$A = (a^2 + b^2)^{\frac{1}{2}} \qquad c = \cos$$

$$B = \frac{1}{2\sqrt{2}} (a + A)^{\frac{1}{2}} \qquad s = \sin$$

$$C = \frac{1}{2\sqrt{2}} (-a + A)^{\frac{1}{2}} \qquad ch = \cosh \qquad sh = \sinh$$

To give the complete expression for a general medium the matrix on the previous page should be multiplied with $\frac{A}{2}\, e^{-Abs}$.

The expression (4) is obviously very complex and very difficult to grasp or use; therefore it is necessary to investigate simplifications.

SIMPLIFICATION 1

Assumption: $LD, LB > CD, CB \quad LD' = LB' = 0$

Consequently: $a \simeq LD^2 - LB^2$, $b \simeq 2LD \cdot LB$

$A = LD^2 + LB^2$, $B = LD/2$, $C = LB/2$

Under these conditions the Mueller matrix looks as given on the next page (common factor e^{-Abs} omitted for convenience).

SIMPLIFICATION 2

Assumption: $CD, CB > LD, LB \quad LD' = LB' = 0$

Consequently: $a \simeq CD^2 - CB^2$, $b \simeq 2CD \cdot CB$

$A = CD^2 + CB^2$, $B = CD/2$, $C = CB/2$

The reader is referred to ref. 8 for the full expression which, however, shows similarities with eq.(5)

$$\begin{pmatrix} \mathrm{chLD} & 2(\mathrm{LD}\cdot\mathrm{CB}-\mathrm{LB}\cdot\mathrm{CD})\cdot(\mathrm{ch}^2\mathrm{B}-\mathrm{c}^2\mathrm{C})/(\mathrm{LD}^2+\mathrm{LB}^2) & (\mathrm{shLD}(\mathrm{LB}\cdot\mathrm{CB}+\mathrm{LD}\cdot\mathrm{CD})+\mathrm{sLB}\cdot(\mathrm{LB}\cdot\mathrm{CD}-\mathrm{LD}\cdot\mathrm{CB}))/(\mathrm{LD}^2+\mathrm{LB}^2) & \mathrm{shLD} \\ 2(\mathrm{LB}\cdot\mathrm{CD}-\mathrm{LD}\cdot\mathrm{CB})\cdot(\mathrm{ch}^2\mathrm{B}-\mathrm{c}^2\mathrm{C})/(\mathrm{LD}^2+\mathrm{LB}^2) & \mathrm{cLB} & \mathrm{sLB} & (\mathrm{shLD}\cdot(\mathrm{LB}\cdot\mathrm{CD}-\mathrm{LD}\cdot\mathrm{CB})+\mathrm{sLB}\cdot(-\mathrm{LB}\cdot\mathrm{CB}-\mathrm{LD}\cdot\mathrm{CD}))/(\mathrm{LD}^2+\mathrm{LB}^2) \\ (\mathrm{shLD}\cdot(\mathrm{LB}\cdot\mathrm{CB}+\mathrm{LD}\cdot\mathrm{CD})+\mathrm{sLB}\cdot(\mathrm{LB}\cdot\mathrm{CD}-\mathrm{LD}\cdot\mathrm{CB}))/(\mathrm{LD}^2+\mathrm{LB}^2) & -\mathrm{sLB} & \mathrm{cLB} & 2(\mathrm{LD}\cdot\mathrm{CD}+\mathrm{LB}\cdot\mathrm{CB})\cdot(\mathrm{ch}^2\mathrm{B}-\mathrm{c}^2\mathrm{C})/(\mathrm{LD}^2+\mathrm{LB}^2) \\ \mathrm{shLD} & (\mathrm{shLD}\cdot(\mathrm{LD}\cdot\mathrm{CB}-\mathrm{LB}\cdot\mathrm{CD})+\mathrm{sLB}\cdot(\mathrm{LB}\cdot\mathrm{CB}+\mathrm{LD}\cdot\mathrm{CD}))/(\mathrm{LD}^2+\mathrm{LB}^2) & 2(\mathrm{LD}\cdot\mathrm{CD}+\mathrm{LB}\cdot\mathrm{CB})\cdot(\mathrm{ch}^2\mathrm{B}-\mathrm{c}^2\mathrm{C})/(\mathrm{LD}^2+\mathrm{LB}^2) & \mathrm{chLD} \end{pmatrix} \quad (5)$$

SIMPLIFICATION 3

We may finally assume that all effects are small so that trigonometric functions may be substituted with power series expansions to the second order; the Mueller matrix then looks as follows:

$$e^{-Abs}\left[\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} - \begin{pmatrix} 0 & -LD' & -CD & -LD \\ -LD' & 0 & -LB & CB \\ -CD & LB & 0 & -LB' \\ -LD & -CB & LB' & 0 \end{pmatrix} \right.$$

$$\left. +\frac{1}{2}\begin{pmatrix} LD^2+LD'^2 & LD\cdot CB- LB\cdot CD & 0 & LB'\cdot CD- LD'\cdot CB \\ LB\cdot CD- LD\cdot CB & LD'^2- LB^2 & LD'\cdot CD+ LB'\cdot CB & LD\cdot LD'+ LB\cdot LB' \\ 0 & LD'\cdot CD+ LB'\cdot CB & -LB^2- LB'^2 & LD\cdot CD+ LB\cdot CB \\ LD'\cdot CB+ LB'\cdot CD & LD'\cdot LD+ LB'\cdot LB & LD\cdot CD+ LB\cdot CB & LD^2- LB'^2 \end{pmatrix}\right] \quad (6)$$

A short-hand for this expression is

$$e^{-Abs}\cdot e^{-\gamma\underline{H}} \quad (7)$$ with

$$\gamma\underline{H} = \begin{pmatrix} 0 & -LD' & -CD & -LD \\ -LD' & 0 & -LB & CB \\ -CD & LB & 0 & -LB' \\ -LD & -CB & LB' & 0 \end{pmatrix}$$

The argument includes a power series expansion to the second order of $e^{-\gamma \underline{H}}$,

$$e^{-\gamma \underline{H}} = \underline{I} - \gamma \underline{H} + (\gamma \underline{H})^2/2$$

evaluation of (LB·LD' - LB'·LD) showing that this is zero, and the acceptance of almost equality between e.g. $(LD^2+LD'^2)$ and $(LD^2+LD'^2+CD^2)$ a question which arises for the diagonal elements in the second order matrix.

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