Planar Laser Polarisation Spectroscopy Imaging in Combustion
Volume II: Appendices

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Appendices

Appendix I: Calculation of the Clebsch-Gordon Coefficient Sums

The algebraic expressions representing the Clebsch-Gordon coefficients used in this thesis are those quoted in Zare *"Angular Momentum"* (p 57, Table 2.4 C: $j_2 = 1$)[A1].

Zare, *"Angular Momentum"*, (p 57), Table 2.4: *"Algebraic expressions for some commonly occurring Clebsch-Gordon Coefficients* $\langle j_1\, m_1, j_2\, m_2 | j, m\rangle$, *Part C: $j_2 = 1$"*

Left circularly polarised transitions

$$\langle j_1\ m-1, 1\ \ 1 | j_1+1\ m\rangle = \sqrt{\frac{(j_1+m)\cdot(j_1+m+1)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}} \qquad \text{Equation 1}$$

$$\langle j_1\ m-1, 1\ \ 1 | j_1\ m\rangle = \sqrt{\frac{(j_1+m)\cdot(j_1-m+1)}{2\cdot j_1\cdot(j_1+1)}} \qquad \text{Equation 2}$$

$$\langle j_1\ m-1, 1\ \ 1 | j_1-1\ \ m\rangle = \sqrt{\frac{(j_1-m)\cdot(j_1-m+1)}{2\cdot j_1\cdot(2\cdot j_1+1)}} \qquad \text{Equation 3}$$

Linearly polarised transitions

$$\langle j_1\ m, 1\ \ 0 | j_1+1\ m\rangle = \sqrt{\frac{(j_1-m+1)\cdot(j_1+m+1)}{(2\cdot j_1+1)\cdot(j_1+1)}} \qquad \text{Equation 4}$$

$$\langle j_1\ m, 1\ \ 0 | j_1\ m\rangle = \frac{m}{\sqrt{j_1\cdot(j_1+1)}} \qquad \text{Equation 5}$$

$$\langle j_1\ m, 1\ \ 0 | j_1-1\ m\rangle = -\sqrt{\frac{(j_1-m)\cdot(j_1+m)}{j_1\cdot(2\cdot j_1+1)}} \qquad \text{Equation 6}$$

Right circularly polarised transitions

$$\langle j_1\ m+1, 1\ \ -1 | j_1+1\ m\rangle = \sqrt{\frac{(j_1-m)\cdot(j_1-m+1)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}} \qquad \text{Equation 7}$$

$$\langle j_1\ m+1, 1\ \ -1 | j_1\ m\rangle = \sqrt{\frac{(j_1-m)\cdot(j_1+m+1)}{2\cdot j_1\cdot(j_1+1)}} \qquad \text{Equation 8}$$

$$\langle j_1\ m+1, 1\ \ -1 | j_1-1\ \ m\rangle = \sqrt{\frac{(j_1+m+1)\cdot(j_1+m)}{2\cdot j_1\cdot(2\cdot j_1+1)}} \qquad \text{Equation 9}$$

The squares of the Clebsch-Gordon coefficients represent probabilities, while the Clebsch-Gordon coefficients themselves represent probability amplitudes. For convenience, Zare's algebraic expressions for the Clebsch-Gordon coefficients in the case of absorption or emission of a photon, $j_2 = 1$, are repeated above.

Note that the selection rules for the above equations require that the combined state magnetic quantum number, m, represents the algebraic sum of the two component magnetic quantum numbers:

$$m = m_1 + m_2 \qquad \text{Equation 10}$$

The combined rotational quantum number, j, is the vector sum of the two component rotational quantum numbers:

$$|j_1 + j_2| \geq j \geq |j_1 - j_2| \qquad \text{Equation 11}$$

This requires that the Clebsch-Gordon coefficients for P ($\Delta j = -1$) transitions are zero for $j_1 = 0$ and ½ and Q ($\Delta j = 0$) transitions are zero for $j_1 = 0$ for the case of absorption or emission of a photon ($j_2 = 1$).

For convenience, we rewrite these equations in terms of the magnetic quantum number of the lower state, m_1, of the transition. The restricted selection rules quoted above are stated directly in the following expressions.

Left circularly polarised transitions ($m_1 = m - 1$, $m = m_1 + 1$)

R transition

$$\langle j_1\ m_1, 1\ 1 \mid j_1{+}1\ m_1{+}1\rangle = \sqrt{\frac{(j_1 + m_1 + 1)\cdot(j_1 + m_1 + 2)}{(2\cdot j_1 + 1)\cdot(2\cdot j_1 + 2)}} \qquad \text{Equation 12}$$

Q transition

$$\langle j_1\ m_1, 1\ 1 \mid j_1\ m_1{+}1\rangle = \mathrm{if}\left[j_1 = 0, 0, -\sqrt{\frac{(j_1 + m_1 + 1)\cdot(j_1 - m_1)}{2\cdot j_1\cdot(j_1 + 1)}}\right] \qquad \text{Equation 13}$$

P transition

$$\langle j_1\ m_1, 1\ 1 \mid j_1{-}1\ m_1{+}1\rangle = \mathrm{if}\left[j_1 < 1, 0, \sqrt{\frac{(j_1 - m_1 - 1)\cdot(j_1 - m_1)}{2\cdot j_1\cdot(2\cdot j_1 + 1)}}\right] \qquad \text{Equation 14}$$

Linearly polarised transitions ($m_1 = m$)

R transition

$$\langle j_1\ m_1, 1\ 0 \mid j_1{+}1\ m_1\rangle = \sqrt{\frac{(j_1 - m_1 + 1)\cdot(j_1 + m_1 + 1)}{(2\cdot j_1 + 1)\cdot(j_1 + 1)}} \qquad \text{Equation 15}$$

Q transition

$$<j_1\ m_1, 1\ \ 0\,|\,j_1\ m_1> = \mathrm{if}\left[j_1 = 0, 0, \frac{m_1}{\sqrt{j_1\cdot(j_1+1)}}\right] \qquad \text{Equation 16}$$

P transition

$$<j_1\ m_1, 1\ \ 0\,|\,j_1\text{-}1\ m_1> = \mathrm{if}\left[j_1 < 1, 0, -\sqrt{\frac{(j_1-m_1)\cdot(j_1+m_1)}{j_1\cdot(2\cdot j_1+1)}}\right] \qquad \text{Equation 17}$$

<u>Right circularly polarised transitions</u> ($m_1 = m + 1$, $m = m_1 - 1$)

R transition

$$<j_1\ m_1, 1\ \ \text{-}1\,|\,j_1\text{+}1\ m_1 - 1> = \sqrt{\frac{(j_1-m_1+1)\cdot(j_1-m_1+2)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}} \qquad \text{Equation 18}$$

Q transition

$$<j_1\ m_1, 1\ \ \text{-}1\,|\,j_1\ m_1 - 1> = \mathrm{if}\left[j_1 = 0, 0, \sqrt{\frac{(j_1-m_1+1)\cdot(j_1+m_1)}{2\cdot j_1\cdot(j_1+1)}}\right] \qquad \text{Equation 19}$$

P transition

$$<j_1\ m_1, 1\ \ \text{-}1\,|\,j_1\text{-}1\ \ m_1 - 1> = \mathrm{if}\left[j_1 < 1, 0, \sqrt{\frac{(j_1+m_1)\cdot(j_1+m_1-1)}{2\cdot j_1\cdot(2\cdot j_1+1)}}\right] \qquad \text{Equation 20}$$

Note that the right circularly polarised Clebsch-Gordon coefficients are equivalent, on replacement of m_1 by $-m_1$, to the left circularly polarised Clebsch-Gordon coefficients.

Average of Right and Left Circularly Polarised Transition Clebsch-Gordon Coefficient Squares

As discussed at the end of this Appendix, Teets, Kowalski, Hill, Carlson and Hansch calculate the probability of absorption of a photon from the orthogonally polarised probe beam component by averaging the squared Clebsch-Gordon coefficients for right and left circularly polarised light. For convenience in the following calculations, we calculate here the average of the right and left circularly polarised Clebsch-Gordon squares.

R transitions

$$\frac{\begin{bmatrix}\langle j_1\ m_1\ 1\ -1|\ j_1+1\ \ m_1-1\rangle \\ +\langle j_1\ m_1\ 1\ +1|\ j_1+1\ \ m_1+1\rangle\end{bmatrix}}{2}=\frac{1}{2}\left\{\left(\frac{(j_1+m_1+1)(j_1+m_1+2)}{(2j_1+1)(2j_1+2)}\right)+\left(\frac{(j_1-m_1+1)(j_1-m_1+2)}{(2j_1+1)(2j_1+2)}\right)\right\}$$

$$\frac{\begin{bmatrix}\langle j_1\ m_1\ 1\ -1|\ j_1+1\ \ m_1-1\rangle \\ +\langle j_1\ m_1\ 1\ +1|\ j_1+1\ \ m_1+1\rangle\end{bmatrix}}{2}=\frac{1}{2}\frac{\left(j_1^2+m_1^2+3j_1+2\right)}{(2j_1+1)(j_1+1)} \qquad \text{Equation 21}$$

Q transitions

$$\frac{\begin{bmatrix}\langle j_1\ m_1\ 1\ -1|\ j_1\ \ m_1-1\rangle \\ +\langle j_1\ m_1\ 1\ +1|\ j_1\ \ m_1+1\rangle\end{bmatrix}}{2}=\frac{1}{2}\left\{\left(\frac{(j_1+m_1+1)(j_1-m_1)}{2j_1(j_1+1)}\right)+\left(\frac{(j_1-m_1+1)(j_1+m_1)}{2j_1(j_1+1)}\right)\right\}$$

$$\frac{\begin{bmatrix}\langle j_1\ m_1\ 1\ -1|\ j_1\ \ m_1-1\rangle \\ +\langle j_1\ m_1\ 1\ +1|\ j_1\ \ m_1+1\rangle\end{bmatrix}}{2}=\frac{1}{2}\frac{\left(j_1^2-m_1^2+j_1\right)}{j_1(j_1+1)} \qquad \text{Equation 22}$$

P transitions

$$\frac{\begin{bmatrix}\langle j_1\ m_1\ 1\ -1|\ j_1-1\ \ m_1-1\rangle \\ +\langle j_1\ m_1\ 1\ +1|\ j_1-1\ \ m_1+1\rangle\end{bmatrix}}{2}=\frac{1}{2}\left\{\left(\frac{(j_1-m_1-1)(j_1-m_1)}{2j_1(2j_1+1)}\right)+\left(\frac{(j_1+m_1)(j_1+m_1-1)}{2j_1(2j_1+1)}\right)\right\}$$

$$\frac{\begin{bmatrix}\langle j_1\ m_1\ 1\ -1|\ j_1-1\ \ m_1-1\rangle \\ +\langle j_1\ m_1\ 1\ +1|\ j_1-1\ \ m_1+1\rangle\end{bmatrix}}{2}=\frac{1}{2}\frac{\left(j_1^2+m_1^2-j_1\right)}{j_1(2j_1+1)} \qquad \text{Equation 23}$$

Calculation of the Absorption Cross-section Summations

To avoid unnecessary subscripts in the following derivations, we represent the initial quantum state, (j_1, m_1), of the transition as (J,M). This is not to be confused with the quantum state of the combined system, (j,m).

The squares of the Clebsch-Gordon coefficients represent rotational state transition probabilities and are written as σ^{i}_{J,J^*,M,M^*}, in this thesis where
i is the polarisation state required for the transition,
(J,M) is the lower quantum state of the transition, and
(J*,M*) represents the upper quantum state of the transition.
The superscript, *, represents either the superscript, ', referring to the upper quantum state of the pump beam transition, or the superscript, ", referring to the upper quantum state of the probe beam transition.

The following sections calculate, firstly, the absorption cross-section summations defined in equations [21] and [22] of Chapter I and, secondly, the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions defined in equations [25], [33] and [53] respectively of Chapter I. The summations are required to calculate the induced dichroism of Teets, Kowalski, Hill, Carlson and Hansch's theory. All calculations for this Appendix were determined using Mathcad Plus 5.0.

Teets σ_{J,J^*} Absorption Cross-section Summations

Left-hand Circularly Polarised Transitions

$$\text{Teets\_}\sigma\text{\_summation ... + left\_circ\_R\_transition} = \frac{C_{J,J+1}}{2\cdot J+1}\cdot\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot C_{J,J+1}$$

Equation 24

$$\text{Teets\_}\sigma\text{\_summation ... + left\_circ\_Q\_transition} = \frac{C_{J,J}}{2\cdot J+1}\cdot\text{if}\left[J=0,0,\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]$$

$$\text{Teets\_}\sigma\text{\_summation ... + left\_circ\_Q\_transition} = \text{if}\left(J=0,0,\frac{1}{3}\cdot C_{J,J}\right)$$

Equation 25

$$\text{Teets\_}\sigma\text{\_summation ... + left\_circ\_P\_transition} = \frac{C_{J,J-1}}{2\cdot J+1}\cdot\text{if}\left[J<1,0,\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]$$

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{left\_circ\_P\_transition} = \text{if}\left[J<1, 0, \frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot C_{J,J-1}\right]$$ Equation 26

Linearly polarised transitions

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{linear\_R\_transition} = \frac{C_{J,J+1}}{2\cdot J+1}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot C_{J,J+1}$$ Equation 27

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{linear\_Q\_transition} = \frac{C_{J,J}}{2\cdot J+1}\cdot\text{if}\left[J=0, 0, \sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\right]$$

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{linear\_Q\_transition} = \text{if}\left(J=0, 0, \frac{1}{3}\cdot C_{J,J+1}\right)$$ Equation 28

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{linear\_P\_transition} = \frac{C_{J,J-1}}{2\cdot J+1}\cdot\text{if}\left[J<1, 0, \sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\right]$$

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{linear\_P\_transition} = \text{if}\left[J<1, 0, \frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot C_{J,J-1}\right]$$ Equation 29

Right-hand Circularly Polarised Transitions

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{right\_circ\_R\_transition} = \frac{C_{J,J+1}}{2\cdot J+1}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot C_{J,J+1}$$

Equation 30

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{right\_circ\_Q\_transition} = \frac{C_{J,J}}{2\cdot J+1}\cdot\text{if}\left[J<1, 0, \sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\right]$$

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{right\_circ\_Q\_transition} = \text{if}\left(J<1, 0, \frac{1}{3}\cdot C_{J,J}\right)$$ Equation 31

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{right\_circ\_P\_transition} = \frac{C_{J,J-1}}{2\cdot J+1}\cdot\text{if}\left[J=0, 0, \sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]$$

$$\text{Teets\_}\sigma\text{\_summation} \ldots + \text{right\_circ\_P\_transition} = \text{if}\left[J=0, 0, \frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot C_{J,J-1}\right]$$ Equation 32

Note that the summation is defined with respect to all Zeeman levels of the lower rotational state of the transition.

Calculation of the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions

The $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions are related to the rotational cross-sections for probe beam components with polarisations parallel and orthogonal to the pump beam polarisation. For the case of a left circularly polarised pump beam, the orthogonal rotational cross-section is the square of the Clebsch-Gordon coefficient for a right circularly polarised probe beam transition. The parallel rotational cross-section is the square of the Clebsch-Gordon coefficient for a left circularly polarised probe beam transition. Here we assume that the pump and probe beams are co-propagating so that a left circularly polarised pump beam has the same sense of rotation as a left circularly polarised probe beam.

In the case of a linearly polarised pump beam, the orthogonal cross-section is assumed to be represented by the average of the squared Clebsch-Gordon coefficients for right and left circularly polarised light.

To avoid excessive complication, the conditions represented by the restricted selection rules defined in equation [11] are discussed after the derivation of the more general expressions below.

Right circularly polarised probe beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta_{J,J+1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ldots\atop +-\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$$\zeta_{J,J+1,J+1} = \frac{3}{2}\cdot\frac{J}{(J+1)}$$ Equation 33

$$Z_{J,J+1,J+1} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J+1}}{C_{J,J+1}\cdot C_{J,J+1}}\cdot\zeta_{J,J+1,J+1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{2}\cdot\frac{J}{(J+1)}\right]$$

$$Z_{J,J+1,J+1} = \frac{1}{6}\cdot\frac{(2\cdot J+3)^2}{(2\cdot J+1)^2}\cdot\frac{J}{(J+1)}$$ Equation 34

R (probe), Q (pump)

$$\zeta_{J,J,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\cdot\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ldots\atop +-\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$$\zeta_{J,J,J+1} = \frac{-3}{2\cdot(J+1)}$$ Equation 35

$$Z_{J,J,J+1} = \frac{\sigma_{J,J}\cdot\sigma_{J,J+1}}{C_{J,J}\cdot C_{J,J+1}}\cdot\zeta_{J,J,J+1} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{2\cdot(J+1)}\right]$$

$$Z_{J,J,J+1} = \frac{-1}{6}\cdot\frac{(2\cdot J+3)}{((2\cdot J+1)\cdot(J+1))}$$ Equation 36

R (probe), P (pump)

$$\zeta_{J,J-1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ \ldots\ + -\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$$\zeta_{J,J-1,J+1} = \frac{-3}{2}$$ Equation 37

$$Z_{J,J-1,J+1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J+1}}{C_{J,J-1}\cdot C_{J,J+1}}\cdot\zeta_{J,J-1,J+1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{2}\right)$$

$$Z_{J,J-1,J+1} = \frac{-1}{6}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)$$ Equation 38

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta_{J,J+1,J}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\ldots+-\left[(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}}$$

$$\zeta_{J,J+1,J}=\frac{-3}{2\cdot(J+1)}$$ Equation 39

$$Z_{J,J+1,J}=\frac{\sigma_{J,J+1}\cdot\sigma_{J,J}}{C_{J,J+1}\cdot C_{J,J}}\cdot\zeta_{J,J+1,J}=\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{-3}{2\cdot(J+1)}\right]$$

$$Z_{J,J+1,J}=\frac{-1}{6}\cdot\frac{(2\cdot J+3)}{((2\cdot J+1)\cdot(J+1))}$$ Equation 40

Q (probe), Q (pump)

$$\zeta_{J,J,J}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\cdot\left[\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\ldots+-(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}}$$

$$\zeta_{J,J,J}=\frac{3}{2\cdot J\cdot(J+1)}$$ Equation 41

$$Z_{J,J,J}=\frac{\sigma_{J,J}\cdot\sigma_{J,J}}{C_{J,J}\cdot C_{J,J}}\cdot\zeta_{J,J,J}=\left(\frac{1}{3}\right)\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{3}{2\cdot J\cdot(J+1)}\right]$$

$$Z_{J,J,J}=\frac{1}{6\cdot J\cdot(J+1)}$$ Equation 42

Q (probe), P (pump)

$$\zeta_{J,J-1,J}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\left[\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\ \dots\ + -(-1)^{2}\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}}$$

$$\zeta_{J,J-1,J}=\frac{3}{2\cdot J}$$ Equation 43

$$Z_{J,J-1,J}=\frac{\sigma_{J,J-1}\cdot\sigma_{J,J}}{C_{J,J-1}\cdot C_{J,J}}\cdot\zeta_{J,J-1,J}=\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{1}{3}\right)\cdot\left(\frac{3}{2\cdot J}\right)$$

$$Z_{J,J-1,J}=\frac{1}{6}\cdot\frac{(2\cdot J-1)}{((2\cdot J+1)\cdot J)}$$ Equation 44

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta_{J,J+1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdots + -\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J+1,J-1} = \frac{-3}{2}$$ Equation 45

$$Z_{J,J+1,J-1} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J-1}}{C_{J,J+1}\cdot C_{J,J-1}}\cdot\zeta_{J,J+1,J-1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{2}\right)$$

$$Z_{J,J+1,J-1} = \frac{-1}{6}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)$$ Equation 46

P (probe), Q (pump)

$$\zeta_{J,J,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\cdot\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdots + -\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J,J-1} = \frac{3}{2\cdot J}$$ Equation 47

$$Z_{J,J,J-1} = \frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta_{J,J,J-1} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{3}{2\cdot J}\right)$$

$$Z_{J,J,J-1} = \frac{1}{6}\cdot\frac{(2\cdot J-1)}{((2\cdot J+1)\cdot J)}$$ Equation 48

P (probe), P (pump)

$$\zeta_{J,J-1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\ldots + -\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J-1,J-1} = \frac{3}{2\cdot J}\cdot(J+1)$$ Equation 49

$$Z_{J,J-1,J-1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J-1}}{C_{J,J-1}\cdot C_{J,J-1}}\cdot\zeta_{J,J-1,J-1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{2\cdot J}\cdot(J+1)\right]$$

$$Z_{J,J-1,J-1} = \frac{1}{6}\cdot\frac{(2\cdot J-1)^2}{\left[(2\cdot J+1)^2\cdot J\right]}\cdot(J+1)$$ Equation 50

For a left circularly polarised pump beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta_{J,J+1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ \cdots\ + -\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$$\zeta_{J,J+1,J+1} = \frac{3}{2}\cdot\frac{J}{(J+1)}$$ Equation 51

$$Z_{J,J+1,J+1} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J+1}}{C_{J,J+1}\cdot C_{J,J+1}}\cdot\zeta_{J,J+1,J+1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{2}\cdot\frac{J}{(J+1)}\right]$$

$$Z_{J,J+1,J+1} = \frac{1}{6}\cdot\frac{(2\cdot J+3)^2}{(2\cdot J+1)^2}\cdot\frac{J}{(J+1)}$$ Equation 52

R (probe), Q (pump)

$$\zeta_{J,J,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\cdot\left[\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ \cdots\ + -\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$$\zeta_{J,J,J+1} = \frac{-3}{2\cdot(J+1)}$$ Equation 53

$$Z_{J,J,J+1} = \frac{\sigma_{J,J}\cdot\sigma_{J,J+1}}{C_{J,J}\cdot C_{J,J+1}}\cdot\zeta_{J,J,J+1} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{2\cdot(J+1)}\right]$$

$$Z_{J,J,J+1} = \frac{-1}{6}\cdot\frac{(2\cdot J+3)}{((2\cdot J+1)\cdot(J+1))}$$ Equation 54

R (probe), P (pump)

$$\zeta_{J,J-1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\left[\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ldots + -\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$$\zeta_{J,J-1,J+1} = \frac{-3}{2}$$ Equation 55

$$Z_{J,J-1,J+1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J+1}}{C_{J,J-1}\cdot C_{J,J+1}}\cdot\zeta_{J,J-1,J+1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{2}\right)$$

$$Z_{J,J-1,J+1} = \frac{-1}{6}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)$$ Equation 56

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta_{J,J+1,J}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\;...\;+-\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}}$$

$$\zeta_{J,J+1,J}=\frac{-3}{2\cdot(J+1)}$$

Equation 57

$$Z_{J,J+1,J}=\frac{\sigma_{J,J+1}\cdot\sigma_{J,J}}{C_{J,J+1}\cdot C_{J,J}}\cdot\zeta_{J,J+1,J}=\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{-3}{2\cdot(J+1)}\right]$$

$$Z_{J,J+1,J}=\frac{-1}{6}\cdot\frac{(2\cdot J+3)}{((2\cdot J+1)\cdot(J+1))}$$

Equation 58

Q (probe), Q (pump)

$$\zeta_{J,J,J}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\cdot\left[(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\;...\;+-\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}}$$

$$\zeta_{J,J,J}=\frac{3}{2\cdot(J+1)\cdot J}$$

Equation 59

$$Z_{J,J,J}=\frac{\sigma_{J,J}\cdot\sigma_{J,J}}{C_{J,J}\cdot C_{J,J}}\cdot\zeta_{J,J,J}=\left(\frac{1}{3}\right)\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{3}{2\cdot(J+1)\cdot J}\right]$$

$$Z_{J,J,J}=\frac{1}{(6\cdot((J+1)\cdot J))}$$

Equation 60

Q (probe), P (pump)

$$\zeta_{J,J-1,J} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\left[(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\ \ldots \atop +-\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}}$$

$$\zeta_{J,J-1,J} = \frac{3}{2\cdot J}$$ Equation 61

$$Z_{J,J-1,J} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J}}{C_{J,J-1}\cdot C_{J,J}}\cdot\zeta_{J,J-1,J} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{1}{3}\right)\cdot\left(\frac{3}{2\cdot J}\right)$$

$$Z_{J,J-1,J} = \frac{1}{6}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)\cdot J}$$ Equation 62

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta_{J,J+1,J-1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\ldots+-\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J+1,J-1}=\frac{-3}{2}\qquad\text{Equation 63}$$

$$Z_{J,J+1,J-1}=\frac{\sigma_{J,J+1}\cdot\sigma_{J,J-1}}{C_{J,J+1}\cdot C_{J,J-1}}\cdot\zeta_{J,J+1,J-1}=\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{2}\right)$$

$$Z_{J,J+1,J-1}=\frac{-1}{6}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)\qquad\text{Equation 64}$$

P (probe), Q (pump)

$$\zeta_{J,J,J-1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\cdot\left[\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\ldots+-\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J,J-1}=\frac{3}{2\cdot J}\qquad\text{Equation 65}$$

$$Z_{J,J,J-1}=\frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta_{J,J,J-1}=\left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{3}{2\cdot J}\right)$$

$$Z_{J,J,J-1}=\frac{1}{6}\cdot\frac{(2\cdot J-1)}{((2\cdot J+1)\cdot J)}\qquad\text{Equation 66}$$

P (probe), P (pump)

$$\zeta_{J,J-1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\left[\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)} \ldots + -\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J-1,J-1} = \frac{3}{2}\cdot\frac{(J+1)}{J}$$

Equation 67

$$Z_{J,J-1,J-1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J-1}}{C_{J,J-1}\cdot C_{J,J-1}}\cdot\zeta_{J,J-1,J-1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{2}\cdot\frac{(J+1)}{J}\right]$$

$$Z_{J,J-1,J-1} = \frac{1}{6}\cdot\frac{(2\cdot J-1)^2}{(2\cdot J+1)^2\cdot J}\cdot(J+1)$$

Equation 68

For a linearly polarised pump beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta_{J,J+1,J+1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J+1,J+1}=\frac{3}{10}\cdot(2\cdot J-1)\cdot\frac{J}{((2\cdot J+3)\cdot(J+1))}$$ Equation 69

$$Z_{J,J+1,J+1}=\frac{\sigma_{J,J+1}\cdot\sigma_{J,J+1}}{C_{J,J+1}\cdot C_{J,J+1}}\cdot\zeta_{J,J+1,J+1}=\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{10}\cdot\frac{J\cdot(2\cdot J-1)}{((2\cdot J+3)\cdot(J+1))}\right]$$

$$Z_{J,J+1,J+1}=\frac{1}{30}\cdot(2\cdot J+3)\cdot(2\cdot J-1)\cdot\frac{J}{\left[(2\cdot J+1)^2\cdot(J+1)\right]}$$ Equation 70

R (probe), Q (pump)

$$\zeta_{J,J,J+1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J,J+1}=\frac{-3}{10}\cdot\frac{(2\cdot J-1)}{(J+1)}$$ Equation 71

$$Z_{J,J,J+1}=\frac{\sigma_{J,J}\cdot\sigma_{J,J+1}}{C_{J,J}\cdot C_{J,J+1}}\cdot\zeta_{J,J,J+1}=\left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{10}\cdot\frac{(2\cdot J-1)}{(J+1)}\right]$$

$$Z_{J,J,J+1}=\frac{-1}{30}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)}$$ Equation 72

R (probe), P (pump)

$$\zeta_{J,J-1,J+1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J-1,J+1}=\frac{3}{10}$$ Equation 73

$$Z_{J,J-1,J+1}=\frac{\sigma_{J,J-1}\cdot\sigma_{J,J+1}}{C_{J,J-1}\cdot C_{J,J+1}}\cdot\zeta_{J,J-1,J+1}=\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{3}{10}\right)$$

$$Z_{J,J-1,J+1}=\frac{1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)$$ Equation 74

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta_{J,J+1,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta_{J,J+1,J}=\frac{-3}{10}\cdot\frac{(2\cdot J-1)}{(J+1)} \qquad \text{Equation 75}$$

$$Z_{J,J+1,J}=\frac{\sigma_{J,J+1}\cdot\sigma_{J,J}}{C_{J,J+1}\cdot C_{J,J}}\cdot\zeta_{J,J+1,J}=\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{-3}{10}\cdot\frac{(2\cdot J-1)}{(J+1)}\right]$$

$$Z_{J,J+1,J}=\frac{-1}{30}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} \qquad \text{Equation 76}$$

Q probe , Q pump

$$\zeta_{J,J,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta_{J,J,J}=\frac{3}{10}\cdot\frac{((2\cdot J+3)\cdot(2\cdot J-1))}{(J\cdot(J+1))} \qquad \text{Equation 77}$$

$$Z_{J,J,J}=\frac{\sigma_{J,J}\cdot\sigma_{J,J}}{C_{J,J}\cdot C_{J,J}}\cdot\zeta_{J,J,J}=\left(\frac{1}{3}\right)\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{3}{10}\cdot\frac{((2\cdot J+3)\cdot(2\cdot J-1))}{(J\cdot(J+1))}\right]$$

$$Z_{J,J,J}=\frac{\sigma_{J,J}\cdot\sigma_{J,J}}{C_{J,J}\cdot C_{J,J}}\cdot\zeta_{J,J,J}=\frac{1}{30}\cdot(2\cdot J+3)\cdot\frac{(2\cdot J-1)}{(J\cdot(J+1))} \qquad \text{Equation 78}$$

Q (probe), P (pump)

$$\zeta_{J,J-1,J} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta_{J,J-1,J} = \frac{-3}{10}\cdot\frac{(2\cdot J+3)}{J} \qquad \text{Equation 79}$$

$$Z_{J,J-1,J} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J}}{C_{J,J-1}\cdot C_{J,J}}\cdot\zeta_{J,J-1,J} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{10}\cdot\frac{(2\cdot J+3)}{J}\right]$$

$$Z_{J,J-1,J} = \frac{-1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J} \qquad \text{Equation 80}$$

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta_{J,J+1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J+1,J-1} = \frac{3}{10}$$ Equation 81

$$Z_{J,J+1,J-1} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J-1}}{C_{J,J+1}\cdot C_{J,J-1}}\cdot\zeta_{J,J+1,J-1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{3}{10}\right)$$

$$Z_{J,J+1,J-1} = \frac{1}{30}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)^2}\cdot(2\cdot J-1)$$ Equation 82

P (probe), Q (pump)

$$\zeta_{J,J,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]\right]}{\displaystyle\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J,J-1} = \frac{-3}{10}\cdot\frac{(2\cdot J+3)}{J}$$ Equation 83

$$Z_{J,J,J-1} = \frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta_{J,J,J-1} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{10}\cdot\frac{(2\cdot J+3)}{J}\right]$$

$$Z_{J,J,J-1} = \frac{-1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J}$$ Equation 84

P (probe), P (pump)

$$\zeta_{J,J-1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]\right]}{\displaystyle\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta_{J,J-1,J-1} = \frac{3}{10}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)} \qquad \text{Equation 85}$$

$$Z_{J,J-1,J-1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J-1}}{C_{J,J-1}\cdot C_{J,J-1}}\cdot\zeta_{J,J-1,J-1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{10}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)}\right]$$

$$Z_{J,J-1,J-1} = \frac{1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)\cdot\frac{(J+1)}{J} \qquad \text{Equation 86}$$

Summary of the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions

We have summarised the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions below. The functions are subject to the previously stated condition, in the case of $j_2 = 1$, that the functions are zero for:

- P transitions of pump and probe beams for J = ½
- P and Q transitions of pump and probe beams for J = 0

where J is the rotational quantum number of the shared lower state of the pump and probe beam transitions.

Note that we have used the convention that $\Delta\alpha_{right\_circ} = \alpha_{right} - \alpha_{left} = -\Delta_{left\_circ}$. The $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions for left and right circularly polarised light are equal as defined.

For a right or left circularly polarised pump beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta_{J,J+1,J+1} = \frac{3}{2} \cdot \frac{J}{(J+1)} \quad \text{Equation 87}$$

$$Z_{J,J+1,J+1} = \frac{1}{6} \cdot \frac{(2 \cdot J + 3)^2}{(2 \cdot J + 1)^2} \cdot \frac{J}{(J+1)} \quad \text{Equation 88}$$

R (probe), Q (pump)

$$\zeta_{J,J,J+1} = \frac{-3}{2 \cdot (J+1)} \quad \text{Equation 89}$$

$$Z_{J,J,J+1} = \frac{-1}{6} \cdot \frac{(2 \cdot J + 3)}{(2 \cdot J + 1) \cdot (J+1)} \quad \text{Equation 90}$$

R (probe), P (pump)

$$\zeta_{J,J-1,J+1} = \frac{-3}{2} \quad \text{Equation 91}$$

$$Z_{J,J-1,J+1} = \frac{-1}{6} \cdot \frac{(2 \cdot J - 1)}{(2 \cdot J + 1)^2} \cdot (2 \cdot J + 3) \quad \text{Equation 92}$$

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump) $$\zeta_{J,J+1,J}=\frac{-3}{2\cdot(J+1)}$$ Equation 93

$$Z_{J,J+1,J}=\frac{-1}{6}\cdot\frac{(2\cdot J+3)}{((2\cdot J+1)\cdot(J+1))}$$ Equation 94

Q (probe), Q (pump) $$\zeta_{J,J,J}=\frac{3}{2\cdot J\cdot(J+1)}$$ Equation 95

$$Z_{J,J,J}=\frac{1}{(6\cdot(J\cdot(J+1)))}$$ Equation 96

Q (probe), P (pump) $$\zeta_{J,J-1,J}=\frac{3}{2\cdot J}$$ Equation 97

$$Z_{J,J-1,J}=\frac{1}{6}\cdot\frac{(2\cdot J-1)}{((2\cdot J+1)\cdot J)}$$ Equation 98

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump) $$\zeta_{J,J+1,J-1}=\frac{-3}{2}$$ Equation 99

$$Z_{J,J+1,J-1}=\frac{-1}{6}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)$$ Equation 100

P (probe), Q (pump) $$\zeta_{J,J,J-1}=\frac{3}{2\cdot J}$$ Equation 101

$$Z_{J,J,J-1}=\frac{1}{6}\cdot\frac{(2\cdot J-1)}{((2\cdot J+1)\cdot J)}$$ Equation 102

P (probe), P (pump) $$\zeta_{J,J-1,J-1}=\frac{3}{2\cdot J}\cdot(J+1)$$ Equation 103

$$Z_{J,J-1,J-1}=\frac{1}{6}\cdot\frac{(2\cdot J-1)^2}{\left[(2\cdot J+1)^2\cdot J\right]}\cdot(J+1)$$ Equation 104

For a linearly polarised pump beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta_{J,J+1,J+1}=\frac{3}{10}\cdot(2\cdot J-1)\cdot\frac{J}{((2\cdot J+3)\cdot(J+1))}$$ Equation 105

$$Z_{J,J+1,J+1}=\frac{1}{30}\cdot(2\cdot J+3)\cdot(2\cdot J-1)\cdot\frac{J}{\left[(2\cdot J+1)^2\cdot(J+1)\right]}$$ Equation 106

R (probe), Q (pump)

$$\zeta_{J,J,J+1}=\frac{-3}{10}\cdot\frac{(2\cdot J-1)}{(J+1)}$$ Equation 107

$$Z_{J,J,J+1}=\frac{-1}{30}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)}$$ Equation 108

R (probe), P (pump)

$$\zeta_{J,J-1,J+1}=\frac{3}{10}$$ Equation 109

$$Z_{J,J-1,J+1}=\frac{1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)$$ Equation 110

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta_{J,J+1,J}=\frac{-3}{10}\cdot\frac{(2\cdot J-1)}{(J+1)}$$ Equation 111

$$Z_{J,J+1,J}=\frac{-1}{30}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)}$$ Equation 112

Q probe , Q pump

$$\zeta_{J,J,J}=\frac{3}{10}\cdot\frac{((2\cdot J+3)\cdot(2\cdot J-1))}{(J\cdot(J+1))}$$ Equation 113

$$Z_{J,J,J}=\frac{1}{30}\cdot(2\cdot J+3)\cdot\frac{(2\cdot J-1)}{(J\cdot(J+1))}$$ Equation 114

Q (probe), P (pump)

$$\zeta_{J,J-1,J}=\frac{-3}{10}\cdot\frac{(2\cdot J+3)}{J}$$ Equation 115

$$Z_{J,J-1,J}=\frac{-1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J}$$ Equation 116

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump) $$\zeta_{J,J+1,J-1}=\frac{3}{10}$$ Equation 117

$$Z_{J,J+1,J-1}=\frac{1}{30}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)^2}\cdot(2\cdot J-1)$$ Equation 118

P (probe), Q (pump) $$\zeta_{J,J,J-1}=\frac{-3}{10}\cdot\frac{(2\cdot J+3)}{J}$$ Equation 119

$$Z_{J,J,J-1}=\frac{-1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J}$$ Equation 120

P (probe), P (pump) $$\zeta_{J,J-1,J-1}=\frac{3}{10}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)}$$ Equation 121

$$Z_{J,J-1,J-1}=\frac{1}{30}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)\cdot\frac{(J+1)}{J}$$ Equation 122

Addendum to Appendix I: The orthogonal Clebsch-Gordon coefficient required for the case of a linearly polarised pump beam for the calculation of the induced linear dichroism

The standard Clebsch-Gordon coefficients tabulated by Zare and listed in Table 1 below can be interpreted as the probability amplitude for the state, (j_f, m_f), to have originated from the combination of the two states, (j_i, m_i) and (1,m) (representing the lower rotational state[1] of the target species and the interacting photon respectively). The m term in the second state represents the polarisation state of the absorbed photon (+1; left-circularly polarised, 0; linearly polarised, -1; right-circularly polarised). The Clebsch-Gordon coefficients listed in Table 1 assume that the quantisation axis lies parallel to the direction of propagation for a circularly polarised pump beam and parallel to the polarisation axis of a linearly polarised pump beam.

The theoretical model described in Chapter II calculates the induced dichroism in terms of sums of squares of Clebsch-Gordon coefficients for probe beam components polarised parallel to and orthogonal to the polarisation direction of the pump beam. In the case of a circularly polarised pump beam (and a co-propagating geometry), the "parallel" and "orthogonal" probe beam components correspond to circularly polarised light of the same and of opposite handedness to the pump beam polarisation respectively. The right and left circular polarisation Clebsch-Gordon coefficients from Table 1 may be used directly in the required summations. However, if we assume the quantisation axis lies parallel to the pump polarisation axis for the case of a linearly polarised pump beam, expressions for the Clebsch-Gordon coefficients for absorption of a photon with polarisation axis normal to the quantisation axis are required to complete the required summations.

The Clebsch-Gordon coefficients of Table 1 are more accurately described as

(i) a set of constants representing the probability amplitude for the final state, (j_f, m_f), to have originated from the combination of the two states, (j_i, m_i) and (1,m), and

(ii) a linked set of (Δm) selection rules characteristic of the transition.

The three component tables in Table 1 represent sets of Clebsch-Gordon coefficients selected by the standard selection rules for absorption of right and left circularly and linearly polarised photons(Δm =1; left-circularly polarised, Δm = 0; linearly polarised, Δm = -1; right-circularly polarised). The selection rules are determined by interpretation of the dipole moment matrix

[1] The coefficients have been rewritten in terms of the lower state of the transition for convenience in describing polarisation spectroscopy.

element for the given geometrical relationship between the quantisation axis and the electric field of the absorbed/emitted photon.

Left circularly polarised transitions ($m_1 = m - 1$, $m = m_1 + 1$)

R transition	$\langle j_1\ m_1, 1\ 1 \mid j_1{+}1\ m_1{+}1\rangle = \sqrt{\dfrac{(j_1+m_1+1)\cdot(j_1+m_1+2)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}}$
Q transition	$\langle j_1\ m_1, 1\ 1 \mid j_1\ m_1{+}1\rangle = \mathrm{if}\left[j_1 = 0, 0, -\sqrt{\dfrac{(j_1+m_1+1)\cdot(j_1-m_1)}{2\cdot j_1\cdot(j_1+1)}}\right]$
P transition	$\langle j_1\ m_1, 1\ 1 \mid j_1{-}1\ m_1{+}1\rangle = \mathrm{if}\left[j_1 < 1, 0, \sqrt{\dfrac{(j_1-m_1-1)\cdot(j_1-m_1)}{2\cdot j_1\cdot(2\cdot j_1+1)}}\right]$

Linearly polarised transitions ($m_1 = m$)

R transition	$\langle j_1\ m_1, 1\ 0 \mid j_1{+}1\ m_1\rangle = \sqrt{\dfrac{(j_1-m_1+1)\cdot(j_1+m_1+1)}{(2\cdot j_1+1)\cdot(j_1+1)}}$
Q transition	$\langle j_1\ m_1, 1\ 0 \mid j_1\ m_1\rangle = \mathrm{if}\left[j_1 = 0, 0, \dfrac{m_1}{\sqrt{j_1\cdot(j_1+1)}}\right]$
P transition	$\langle j_1\ m_1, 1\ 0 \mid j_1{-}1\ m_1\rangle = \mathrm{if}\left[j_1 < 1, 0, -\sqrt{\dfrac{(j_1-m_1)\cdot(j_1+m_1)}{j_1\cdot(2\cdot j_1+1)}}\right]$

Right circularly polarised transitions ($m_1 = m + 1$, $m = m_1 - 1$)

R transition	$\langle j_1\ m_1, 1\ {-1} \mid j_1{+}1\ m_1-1\rangle = \sqrt{\dfrac{(j_1-m_1+1)\cdot(j_1-m_1+2)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}}$
Q transition	$\langle j_1\ m_1, 1\ {-1} \mid j_1\ m_1-1\rangle = \mathrm{if}\left[j_1 = 0, 0, \sqrt{\dfrac{(j_1-m_1+1)\cdot(j_1+m_1)}{2\cdot j_1\cdot(j_1+1)}}\right]$
P transition	$\langle j_1\ m_1, 1\ {-1} \mid j_1{-}1\ m_1-1\rangle = \mathrm{if}\left[j_1 < 1, 0, \sqrt{\dfrac{(j_1+m_1)\cdot(j_1+m_1-1)}{2\cdot j_1\cdot(2\cdot j_1+1)}}\right]$

Table 1: Clebsch-Gordon coefficients listed in Zare[A1] rewritten in terms of the rotational and magnetic quantum numbers of the lower state, j_1 and m_1.

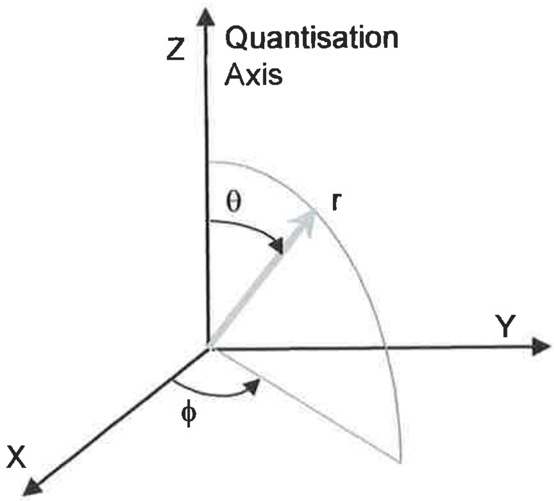

Figure 1: Convention for the spherical co-ordinates, (r,θ,ϕ), for this addendum.

In order to determine the required selection rules for the "orthogonal" Clebsch-Gordon coefficients in the case of a linearly polarised pump beam, we consider a geometry with the usual spherical co-ordinates, (r,θ,ϕ), as shown in Figure 1 above. The rectangular Cartesian vector description of the position vector, $\underline{r}$, is written

$$\underline{r} = \begin{pmatrix} r\sin(\theta)\cos(\phi) \\ r\sin(\theta)\sin(\phi) \\ r\cos(\theta) \end{pmatrix}$$ Equation 1

where the Z axis is the quantisation axis for the magnetic quantum number.

We can also write vectors representing the electric field (ignoring the common time dependent component) of a probe beam component for four geometries:

Left circularly polarised probe beam component (m = 1 with respect to the Z quantisation axis)

$$\underline{E}_{left-hand} = E_0 \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix}$$ Equation 2

Right circularly polarised probe beam component (m = -1 with respect to the Z quantisation axis)

$$\underline{E}_{right-hand} = E_0 \begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix}$$ Equation 3

Linearly polarised probe beam component polarised parallel to the Z axis (m = 0 with respect to the Z quantisation axis)

$$\underline{E}_{linear\_z} = E_0 \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \quad \text{Equation 4}$$

Linearly polarised probe beam component polarised parallel to the X axis

$$\underline{E}_{linear\_x} = E_0 \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \quad \text{Equation 5}$$

The resultant scalar product of the position and electric field vectors in each case are

<u>Left circularly polarised probe beam component</u> ($\Delta m = 1$ with respect to the Z quantisation axis)

$$\underline{r} \cdot \underline{E}_{left\text{-}hand} = rE_0 \sin(\theta)(\cos(\phi) + i\sin(\phi)) = rE_0 \sin(\theta) e^{i\phi} \quad \text{Equation 6}$$

<u>Right circularly polarised probe beam component</u> ($\Delta m = -1$ with respect to the Z quantisation axis)

$$\underline{r} \cdot \underline{E}_{right\text{-}hand} = rE_0 \sin(\theta)(\cos(\phi) - i\sin(\phi)) = rE_0 \sin(\theta) e^{-i\phi} \quad \text{Equation 7}$$

<u>Linearly polarised probe beam component polarised parallel to the Z axis</u> ($\Delta m = 0$ with respect to the Z quantisation axis)

$$\underline{r} \cdot \underline{E}_{linear\_z} = rE_0 \cos(\theta) \quad \text{Equation 8}$$

<u>Linearly polarised probe beam component polarised parallel to the X axis</u>

$$\underline{r} \cdot \underline{E}_{linear\_x} = rE_0 \sin(\theta)\cos(\phi) \quad \text{Equation 9}$$

The dipole matrix element, D, may be written in separable terms of the variables of the spherical coordinate system as the function describing the state of the system is given by [A2]

$$\Psi_{njm}(r,\theta,\phi) = R_{nj}(r)\Theta_{jm}(\theta)\Phi_m(\phi) \quad \text{Equation 10}$$

where, from normalisation,

$$\int_0^{2\pi} \Phi_m^*(\phi)\Phi_m(\phi)\, d\phi = 1 \quad \text{Equation 11}$$

$$\int_0^{\pi} \Theta_{jm}^*(\theta)\Theta_{jm}(\theta)\sin(\theta)\, d\theta = 1 \quad \text{Equation 12}$$

$$\int_0^{\infty} R_{nj}^*(r)R_{nj}(r)r^2\, dr = 1 \quad \text{Equation 13}$$

and the spherical harmonic components of the state function are

$$\Phi_m(\phi) = \frac{1}{\sqrt{2\pi}} e^{im\phi} \quad \text{Equation 14}$$

$$\Theta_{jm}(\theta) = \sqrt{\frac{(2j+1)(j-m)!}{2(j+m)!}}\, P_j^m(\cos(\theta)) \quad \text{Equation 15}$$

where $P_j^m(\cos(\theta))$ is the associated Legendre function.

The dipole matrix element in the four polarisation geometries considered above separate into the following separable integrals which lead to the appropriate selection rules in each case

Left circularly polarised probe beam component ($\Delta m = 1$ with respect to the Z quantisation axis)

$$D_{left-hand} = eE_0 \int_0^{\infty} R_{nj}^*(r) R_{nj}(r) r^3 \, dr \int_0^{\pi} \Theta_{jm}^*(\theta) \Theta_{jm}(\theta) \sin^2(\theta) d\theta \int_0^{2\pi} \Phi_m^*(\phi) \Phi_m(\phi) e^{i\phi} d\phi \qquad \text{Equation 16}$$

Right circularly polarised probe beam component ($\Delta m = -1$ with respect to the Z quantisation axis)

$$D_{right-hand} = eE_0 \int_0^{\infty} R_{nj}^*(r) R_{nj}(r) r^3 \, dr \int_0^{\pi} \Theta_{jm}^*(\theta) \Theta_{jm}(\theta) \sin^2(\theta) d\theta \int_0^{2\pi} \Phi_m^*(\phi) \Phi_m(\phi) e^{-i\phi} d\phi \qquad \text{Equation 17}$$

Linearly polarised probe beam component polarised parallel to the Z axis ($\Delta m = 0$ with respect to the Z quantisation axis)

$$D_{linear\_z} = eE_0 \int_0^{\infty} R_{nj}^*(r) R_{nj}(r) r^3 \, dr \int_0^{\pi} \Theta_{jm}^*(\theta) \Theta_{jm}(\theta) \sin(\theta)\cos(\theta) d\theta \int_0^{2\pi} \Phi_m^*(\phi) \Phi_m(\phi) d\phi \qquad \text{Equation 18}$$

Linearly polarised probe beam component polarised parallel to the X axis

$$D_{linear\_x} = eE_0 \int_0^{\infty} R_{nj}^*(r) R_{nj}(r) r^3 \, dr \int_0^{\pi} \Theta_{jm}^*(\theta) \Theta_{jm}(\theta) \sin^2(\theta) d\theta \int_0^{2\pi} \Phi_m^*(\phi) \Phi_m(\phi) \cos(\phi) d\phi \qquad \text{Equation 19}$$

Considering only the integrals corresponding to the spherical harmonics, we can see that the selection rule for the magnetic quantum number for the linearly polarised probe beam component polarised along the quantisation (Z) axis is $\Delta m = 0$. It can also be seen that the selection rules for the circularly polarised probe beam components, $\Delta m = 1$ and $\Delta m = -1$ for left and right circularly polarised components respectively, are shared by the probe beam component polarised normal to the quantisation axis, $\Delta m = \pm 1$, in the case of a linearly polarised pump beam. It is clear that the electric field in the last mentioned case can be written as linear combination of right and left circularly polarised components, as can the dipole moment matrix element leading to this selection rule. The equivalent table of Clebsch-Gordon coefficients to Table 1 for the "orthogonal" probe beam component in the case of a linearly polarised pump beam given this selection rule is given below as Table 2.

The implication of the selection rule for the linearly polarised probe beam component polarised normal to the quantisation axis in the case of a linearly polarised pump beam is that Teets, Kowalski, Hill, Carlson and Hansch calculate the absorption of this component as the average of the transition probabilities (proportional to the squares of the Clebsch-Gordon coefficients) for the cases of left and right circularly polarised pump photons. The calculation of the induced dichroism according to equations [6] to [8] of Chapter I is now explicitly a sum over the possible absorption routes allowed by the selection rules and weighted by the probability of each transition (i.e. by the square of the Clebsch-Gordon coefficients). For the case of a linearly polarised pump beam, equation [6] of Chapter I now becomes

$$\zeta_{J,J',J''}^{\substack{linear\_pump \\ \_same\_electronic\_state \\ and\_same\_vibrational\_manifold}} = (2J+1)\cdot\frac{\sum_{M}\sigma^{i_{pump}}_{J,J',M,M'}\cdot\left(\sigma^{linear\_z}_{J,J'',M,M''}-\left(\frac{\sigma^{left\_circ}_{J,J'',M,M''}+\sigma^{right\_circ}_{J,J'',M,M''}}{2}\right)\right)}{\sum_{M}\sigma^{i_{pump}}_{J,J',M,M'}\cdot\sum_{M}\sigma^{i_{probe}}_{J,J'',M,M''}} \qquad \text{Equation 20}$$

Transitions for the case of a linearly polarised probe beam component polarised normal to the quantisation axis ($\Delta m = \pm 1$)

R transitions	$\langle j_1\ m_1, 1\ -1 \mid j_1+1\ m_1-1\rangle = \sqrt{\frac{(j_1-m_1+1)\cdot(j_1-m_1+2)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}}$ $\langle j_1\ m_1, 1\ 1 \mid j_1+1\ m_1+1\rangle = \sqrt{\frac{(j_1+m_1+1)\cdot(j_1+m_1+2)}{(2\cdot j_1+1)\cdot(2\cdot j_1+2)}}$
Q transitions	$\langle j_1\ m_1, 1\ -1 \mid j_1\ m_1-1\rangle = \mathrm{if}\left[j_1=0, 0, \sqrt{\frac{(j_1-m_1+1)\cdot(j_1+m_1)}{2\cdot j_1\cdot(j_1+1)}}\right]$ $\langle j_1\ m_1, 1\ 1 \mid j_1\ m_1+1\rangle = \mathrm{if}\left[j_1=0, 0, -\sqrt{\frac{(j_1+m_1+1)\cdot(j_1-m_1)}{2\cdot j_1\cdot(j_1+1)}}\right]$
P transitions	$\langle j_1\ m_1, 1\ -1 \mid j_1-1\ m_1-1\rangle = \mathrm{if}\left[j_1<1, 0, \sqrt{\frac{(j_1+m_1)\cdot(j_1+m_1-1)}{2\cdot j_1\cdot(2\cdot j_1+1)}}\right]$ $\langle j_1\ m_1, 1\ 1 \mid j_1-1\ m_1+1\rangle = \mathrm{if}\left[j_1<1, 0, \sqrt{\frac{(j_1-m_1-1)\cdot(j_1-m_1)}{2\cdot j_1\cdot(2\cdot j_1+1)}}\right]$

Table 2: Clebsch-Gordon coefficients rewritten in terms of the rotational and magnetic quantum numbers of the lower state, j_1 and m_1, for the case of a linearly polarised probe beam component (which can be considered as a linear combination of right and left circularly polarised probe beam components) polarised normal to the quantisation axis.

References:

[A1] Zare, R.N., "Angular Momentum. Understanding Spatial Aspects in Chemistry and Physics.", John Wiley and Sons, Inc., New York, 1st Ed., 1998.

[A2] Cassels, J.M., "Basic Quantum Mechanics", (p 58 and following). McGraw-Hill, London. New York. Sydney. Toronto. Mexico. Johannesburg. Panama. Singapore. 1970.

Appendix II: Closed Two-level Rate Equation Model

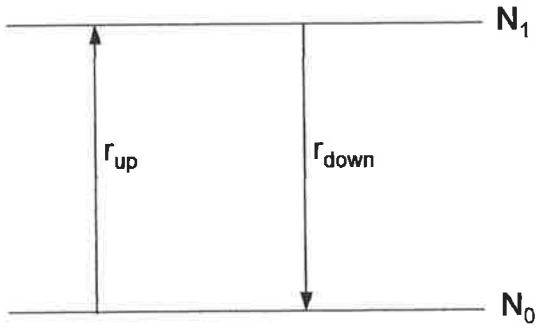

Figure 1: Closed two-level system.

Consider a closed two-level system as shown in the figure above, with lower state population density, N_0, and upper state population density, N_1. The transition rate from the lower to the upper state may include both stimulated absorption and collisional transition rates and is denoted by the term, r_{up}. The transition rate from the upper to the lower state includes spontaneous and stimulated emission as well as collisional transition rates and is denoted by the term, r_{down}. The total transition rate, r, is defined as

$r = r_{up} + r_{down}$. Equation 1

The time dependence of the population density, $N(t)_0$, where t represents time, is given by the rate equation

$$\frac{d}{dt}N(t)_0 = -r_{up} \cdot N(t)_0 + r_{down} \cdot N(t)_1$$ Equation 2

which is equivalent to the equation

$$\frac{d}{dt}N(t)_0 = \left[-r_{up} \cdot N(t)_0 + r_{down} \cdot \left(N - N(t)_0\right)\right] = -r \cdot N(t)_0 + r_{down} \cdot N$$ Equation 3

where N is the total population density defined as

$N = N(t)_0 + N(t)_1$ Equation 4

Equation [3] may be rewritten as the inhomogeneous equation

$$\frac{d}{dt}N(t)_0 + r \cdot N(t)_0 = r_{down} \cdot N$$ Equation 5

The homogeneous solution is

$$N(t)_{0\_homogeneous} = N(0)_0 \cdot e^{-\int_0^t r\,d\tau} = N(0)_0 \cdot e^{-r \cdot t}$$ Equation 6

The particular solution, since the homogeneous solution is non-zero if $N(0)_0$ is non-zero, is given by

$$N(t)_{0\_particular} = \left[1 + \int_0^t \frac{r_{down} \cdot N}{N(0)_0 \cdot e^{-r \cdot \tau}} d\tau = 1 + \frac{r_{down} \cdot N}{r \cdot N(0)_0} \cdot \left(e^{r \cdot t} - 1\right)\right] = 1 + \frac{r_{down} \cdot N}{r \cdot N(0)_0} \cdot \left(e^{r \cdot t} - 1\right)$$

Equation 7

We combine these two solutions to give the general solution

$$N(t)_0 = \left[N(t)_{0\_homogeneous} \cdot N(t)_{0\_particular} = N(0)_0 \cdot e^{-r \cdot t} \cdot \left[1 + \frac{r_{down} \cdot N}{r \cdot N(0)_0} \cdot \left(e^{r \cdot t} - 1\right)\right]\right]$$

or

$$N(t)_0 = N(0)_0 \cdot e^{-r \cdot t} + \frac{r_{down}}{r} \cdot N \cdot \left(1 - e^{-r \cdot t}\right)$$ Equation 8

The population density of the upper state is then given by the equation

$$N(t)_1 = \left(N - N(t)_0\right) = N - N(0)_0 \cdot e^{-r \cdot t} - \frac{r_{down}}{r} \cdot N \cdot \left(1 - e^{-r \cdot t}\right)$$ Equation 9

and the population density difference between the lower and upper states by

$$N(t)_0 - N(t)_1 = \left[N(t)_0 - \left(N - N(t)_0\right) = 2 \cdot N(t)_0 - N\right] = 2 \cdot N(0)_0 \cdot e^{-r \cdot t} + 2 \cdot \frac{r_{down}}{r} \cdot N \cdot \left(1 - e^{-r \cdot t}\right) - N$$

Equation 10

Equations [8] to [10] describe the general population densities for the lower and upper states, and the population density difference between the upper and lower states. We now obtain solutions for the case of an initially unpopulated upper states, where

$N(0)_1 = 0$ and $N(0)_0 = N$ Equation 11

Equations [8] to [10] become

$$N(t)_{0\ ...\ + unpopulated\_upper\_state} = \left[N \cdot e^{-r \cdot t} + \frac{r_{down}}{r} \cdot N \cdot \left(1 - e^{-r \cdot t}\right)\right] = N \cdot \left[1 - \left(\frac{r_{up}}{r}\right) \cdot \left(1 - e^{-r \cdot t}\right)\right]$$

Equation 12

$$N(t)_{1 \ldots + unpopulated\_upper\_state} = \left[N - N \cdot e^{-r \cdot t} - \frac{r_{down}}{r} \cdot N \cdot \left(1 - e^{-r \cdot t}\right) \right] = N \cdot \left(\frac{r_{up}}{r}\right) \cdot \left(1 - e^{-r \cdot t}\right)$$

Equation 13

$$\left(N(t)_0 - N(t)_1\right)_{unpopulated\_upper\_state} = 2 \cdot N \cdot e^{-r \cdot t} \ldots + 2 \cdot \frac{r_{down}}{r} \cdot N \cdot \left(1 - e^{-r \cdot t}\right) - N = N \cdot \left[1 - 2 \cdot \left(\frac{r_{up}}{r}\right) \cdot \left(1 - e^{-r \cdot t}\right)\right]$$

Equation 14

For ease of calculation of the steady-state solutions, we may rewrite equations [12] to [14] as

$$N(t)_{0 \ldots + unpopulated\_upper\_state} = N \cdot \left[\left(\frac{r_{down}}{r}\right) + \left(\frac{r_{up}}{r}\right) \cdot \left(e^{-r \cdot t}\right)\right]$$ Equation 15

$$N(t)_{1 \ldots + unpopulated\_upper\_state} = N \cdot \left[\left(\frac{r_{up}}{r}\right) - \left(\frac{r_{up}}{r}\right) \cdot e^{-r \cdot t}\right]$$ Equation 16

$$\left(N(t)_0 - N(t)_1\right)_{unpopulated\_upper\_state} = N \cdot \left[\left(\frac{r_{down}}{r}\right) - \left(\frac{r_{up}}{r}\right) + 2 \cdot \left(\frac{r_{up}}{r}\right) \cdot e^{-r \cdot t}\right]$$ Equation 17

Equations [15] to [17] give the three population density equations in the case of an initially, t = 0, unpopulated lower state. We now investigate two limiting cases of this solution; for small timescales with respect to the exponential terms of the solutions, rt << 1, where the exponential function may be approximated linearly, and in the steady state regime, rt >> 1.

In the linear regime, rt << 1, the following approximations may be made

$$e^{-r \cdot t} = 1 - r \cdot t$$ Equation 18

and

$$1 - e^{-r \cdot t} = r \cdot t$$ Equation 19

The population density equations in the case of an initially unpopulated upper state in the linear regime become

$$N(t)_{0 \ldots + unpopulated\_upper\_state \ldots + linear\_regime} = \left[N \cdot \left[1 - \left(\frac{r_{up}}{r}\right) \cdot (r \cdot t)\right]\right] = N \cdot \left(1 - r_{up} \cdot t\right)$$ Equation 20

$$N(t)_{1\ \ldots + unpopulated\_upper\_state + linear\_regime} = \left[N\cdot\left(\frac{r_{up}}{r}\right)\cdot(r\cdot t)\right] = N\cdot(r_{up}\cdot t)$$ Equation 21

$$\left(N(t)_0 - N(t)_1\right)_{unpopulated\_upper\_state\ \ldots + linear\_regime} = \left[N\cdot\left[1 - 2\cdot\left(\frac{r_{up}}{r}\right)\cdot(r\cdot t)\right]\right] = N\cdot(1 - 2\cdot r_{up}\cdot t)$$ Equation 22

The steady state solutions, where $e^{-rt} = 0$, in the case of an initially, $t = 0$, unpopulated upper state are

$$N(t)_{0\ \ldots + unpopulated\_upper\_state\ \ldots + steady\_state} = N\cdot\left(\frac{r_{down}}{r}\right)$$ Equation 23

$$N(t)_{1\ \ldots + unpopulated\_upper\_state\ \ldots + steady\_state} = N\cdot\left(\frac{r_{up}}{r}\right)$$ Equation 24

$$\left(N(t)_0 - N(t)_1\right)_{unpopulated\_upper\_state\ \ldots + steady\_state} = N\cdot\left(\frac{r_{down} - r_{up}}{r}\right)$$ Equation 25

If we explicitly state the transition rates in terms for the spontaneous, stimulated and collisional transition rates, denoted A, BW and Q respectively, we may investigate different experimental ranges. Typically the terms r_{up}, r_{down} and r are represented by equations of the form

$$r_{up} = B_{01}\cdot W + Q_{01}$$ Equation 26

$$r_{down} = A_{10} + B_{10}\cdot W + Q_{10}$$ Equation 27

and

$$r = A_{10} + B_{10}\cdot W + B_{01}\cdot W + Q_{10} + Q_{01}$$ Equation 28

where the subscripts indicate the direction of the transition.

The linear regime solutions, in the case of an initially ($t = 0$) unpopulated upper state become

$$N(t)_{0\ \ldots + unpopulated\_upper\_state\ \ldots + linear\_regime} = N\cdot\left[1 - \left(B_{01}\cdot W + Q_{01}\right)\cdot t\right]$$ Equation 29

$$N(t)_{1\ \ldots + unpopulated\_upper\_state\ \ldots + linear\_regime} = N\cdot\left[\left(B_{01}\cdot W + Q_{01}\right)\cdot t\right]$$ Equation 30

$$\left(N(t)_0 - N(t)_1\right)_{unpopulated\_upper\_state\ \ldots + linear\_regime} = N\cdot\left[1 - 2\cdot\left(B_{01}\cdot W + Q_{01}\right)\cdot t\right]$$ Equation 31

For the case of negligible collisional transfer rates from the lower to the upper state populations, $Q_{01} << B_{01}$ W, equations [29] to [31] become

$$N(t)_{0\ \ldots\ +unpopulated\_upper\_state\ \ldots\ +linear\_regime\ \ldots\ +negligible\_upwards\_collisional\_tranfer} = N\cdot(1 - B_{01}\cdot W\cdot t) \quad \text{Equation 32}$$

$$N(t)_{0\ \ldots\ +unpopulated\_upper\_state\ \ldots\ +linear\_regime\ \ldots\ +negligible\_upwards\_collisional\_tranfer} = N\cdot(B_{01}\cdot W\cdot t) \quad \text{Equation 33}$$

$$(N(t)_0 - N(t)_1)_{unpopulated\_upper\_state\ \ldots\ +linear\_regime\ \ldots\ +negligible\_upwards\_collisional\_tranfer} = N\cdot(1 - 2\cdot B_{01}\cdot W\cdot t) \quad \text{Equation 34}$$

The equivalent steady-state solutions do not simplify significantly unless the upper and lower states are of equal degeneracy so that $B_{01} = B_{10}$.

Appendix III: Projections of Complex Vectors

For real vectors, a and b, the inner or dot product is given by

$$a \cdot b = \begin{bmatrix} a_1 \\ a_2 \\ a_3 \end{bmatrix} \cdot \begin{bmatrix} b_1 \\ b_2 \\ b_3 \end{bmatrix} = a_1 \cdot b_1 + a_2 \cdot b_2 + a_3 \cdot b_3 = |a| \cdot |b| \cdot \cos(\theta) \qquad \text{Equation 1}$$

where θ is the angle between the two vectors. Note that as

$$\cos(-\theta) = \cos(\theta) \qquad \text{Equation 2}$$

the inner product provides no information about the sign of the angle, θ, between the two vectors.

In the case of complex vectors, c and d, the inner or dot product is defined as

$$c \cdot d = \begin{bmatrix} c_1 \\ c_2 \\ c_3 \end{bmatrix} \cdot \begin{bmatrix} d_1 \\ d_2 \\ d_3 \end{bmatrix} = c_1 \cdot \left(\overline{d_1}\right) + c_2 \cdot \left(\overline{d_2}\right) + c_3 \cdot \left(\overline{d_3}\right) = |c| \cdot |d| \cdot \cos(\phi) \qquad \text{Equation 3}$$

where the line over the components of the vector, d, is used to indicate a complex conjugate. Note also that this definition allows the angle, ϕ, to be complex.

<u>The complex dot product is not multiplicatively commutative</u>, i.e. the order of the two vectors is not interchangeable. If we <u>define</u> the vector, d, to be a unit vector, the inner or dot product in equation [2] indicates the complex "projection" of the vector, c, onto the vector, d.

Consider the case of two orthogonal unit vectors

$$g = \frac{1}{\sqrt{2}} \cdot \begin{bmatrix} 1 \\ i \\ 0 \end{bmatrix} \quad \text{and} \quad h = \frac{1}{\sqrt{2}} \cdot \begin{bmatrix} 1 \\ -i \\ 0 \end{bmatrix} \qquad \text{Equation 4}$$

representing left and right handed polarisations of light respectively for a beam travelling in the position Z direction.

If we wish to decompose the vector, k, which lies in the XY plane into components parallel and perpendicular to the unit vectors, we write the vector equation

$$k = (k.g)\, g + (k.h)\, h \qquad \text{Equation 5}$$

i.e.

$$k = \begin{bmatrix} k_1 \\ k_2 \\ 0 \end{bmatrix} = \left[\begin{bmatrix} k_1 \\ k_2 \\ 0 \end{bmatrix} \cdot \left[\frac{1}{\sqrt{2}} \cdot \begin{bmatrix} 1 \\ i \\ 0 \end{bmatrix}\right]\right] \cdot \left[\frac{1}{\sqrt{2}} \cdot \begin{bmatrix} 1 \\ i \\ 0 \end{bmatrix}\right] + \left[\begin{bmatrix} k_1 \\ k_2 \\ 0 \end{bmatrix} \cdot \left[\frac{1}{\sqrt{2}} \cdot \begin{bmatrix} 1 \\ -i \\ 0 \end{bmatrix}\right]\right] \cdot \left[\frac{1}{\sqrt{2}} \cdot \begin{bmatrix} 1 \\ -i \\ 0 \end{bmatrix}\right]$$

$$k = \frac{1}{2}\cdot(k_1 - i\cdot k_2)\cdot\begin{bmatrix}1\\ i\\ 0\end{bmatrix} + \frac{1}{2}\cdot(k_1 + i\cdot k_2)\cdot\begin{bmatrix}1\\ -i\\ 0\end{bmatrix}$$ Equation 6

which, when recomposed, recreates the original vector.

If we use the reverse order of vector inner product, i.e.

$k_{reverse\_order} = (g.k)\ g + (h.k)\ h$ Equation 7

we find

$$k_{reverse\_order} = \left[\left[\frac{1}{\sqrt{2}}\cdot\begin{bmatrix}1\\ i\\ 0\end{bmatrix}\right]\cdot\begin{bmatrix}k_1\\ k_2\\ 0\end{bmatrix}\right]\cdot\left[\frac{1}{\sqrt{2}}\cdot\begin{bmatrix}1\\ i\\ 0\end{bmatrix}\right] + \left[\left[\frac{1}{\sqrt{2}}\cdot\begin{bmatrix}1\\ -i\\ 0\end{bmatrix}\right]\cdot\begin{bmatrix}k_1\\ k_2\\ 0\end{bmatrix}\right]\cdot\left[\frac{1}{\sqrt{2}}\cdot\begin{bmatrix}1\\ -i\\ 0\end{bmatrix}\right]$$

$$k_{reverse\_order} = \frac{(k_1 + i\cdot k_2)\cdot\begin{bmatrix}1\\ i\\ 0\end{bmatrix} + (k_1 - i\cdot k_2)\cdot\begin{bmatrix}1\\ -i\\ 0\end{bmatrix}}{2}$$

$$k_{reverse\_order} = \begin{bmatrix}k_1\\ -k_2\\ 0\end{bmatrix}$$ Equation 8

i.e. the vector equation $k_{reverse\_order} = (g.k)\ g + (h.k)\ h$ does not return the input vector on reconstruction.

In conclusion, to decompose a vector, k, into components parallel to a basis set of orthogonal unit vectors, g, h ... , we must use the following order sensitive equation

k = (k.g) g + (k.h) h + ... Equation 9

for definition of the inner or dot product for two unit vectors, c and d, as

$c.d = c_1\cdot\bar{d}_1 + c_2\cdot\bar{d}_2 + c_3\cdot\bar{d}_3$ Equation 10

Note that if we used the opposite order convention for the inner or dot product, i.e.

$c.d = \bar{c}_1\cdot d_1 + \bar{c}_2\cdot d_2 + \bar{c}_3\cdot d_3$ Equation 11

the vector component order in the decomposition equation must be reversed

k = (g.k) g + (h.k) h + ... Equation 12

Appendix IV: First Order Approximation to the Geometric Dependence of the Induced Linear Birefringence

Consider a coordinate system with probe beam propagating along the positive Z axis, i.e.

$$probe_{propagation} = \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix} \qquad \text{Equation 1}$$

The probe beam propagates at the angle, φ, to the optic axis, experiencing a birefringence between extraordinary and ordinary rays, defined in equation [32] of Chapter II as

$$\Delta n = n(\varphi)_e - n_o = \sqrt{\frac{(n_o)^2 \cdot (n_e)^2}{(n_o)^2 \cdot \sin(\varphi)^2 + (n_e)^2 \cdot \cos(\varphi)^2}} - n_o \qquad \text{Equation 2}$$

which may be rearranged as

$$n(\varphi)_e - n_o = \sqrt{\frac{(n_o)^2}{\cos(\varphi)^2 + \left(\frac{n_o}{n_e}\right)^2 \cdot \sin(\varphi)^2}} - n_o$$

$$n(\varphi)_e - n_o = n_o \cdot \sqrt{\frac{1}{\cos(\varphi)^2 + \left(\frac{n_o}{n_e}\right)^2 \cdot \sin(\varphi)^2}} - n_o \qquad \text{Equation 3}$$

If the induced dichroism and birefringence are small, so that

$$\alpha_o = \alpha + \frac{\Delta\alpha}{2} \qquad k_o = k + \frac{\Delta k}{2} \quad \text{and} \quad n_o = n + \frac{\Delta n}{2}$$

$$\alpha_e = \alpha - \frac{\Delta\alpha}{2} \qquad k_e = k - \frac{\Delta k}{2} \quad \text{and} \quad n_e = n - \frac{\Delta n}{2} \qquad \text{Equation 4}$$

we can approximate the fraction

$$\frac{n_e}{n_o} = \frac{n - \frac{\Delta n}{2}}{n + \frac{\Delta n}{2}} = \frac{1 - \frac{\Delta n}{2 \cdot n}}{1 + \frac{\Delta n}{2 \cdot n}} = \left(1 - \frac{\Delta n}{2 \cdot n}\right) \cdot \left(1 - \frac{\Delta n}{2 \cdot n}\right) = 1 - \frac{\Delta n}{n} + \frac{1}{4} \cdot \frac{\Delta n^2}{n^2} \qquad \text{Equation 5}$$

to first order by

$$\frac{n_e}{n_o} = 1 - \frac{\Delta n}{n} \quad \text{i.e.} \quad \frac{\Delta n}{n} = 1 - \frac{n_e}{n_o} \qquad \text{Equation 6}$$

and $\frac{n_o}{n_e} = 1 + \frac{\Delta n}{n}$ i.e. $\frac{\Delta n}{n} = \frac{n_o}{n_e} - 1$ Equation 7

so that, to first order again

$$\left(\frac{n_e}{n_o}\right)^2 = 1 - 2\cdot\frac{\Delta n}{n}$$ Equation 8

and

$$\left(\frac{n_o}{n_e}\right)^2 = 1 + 2\cdot\frac{\Delta n}{n}$$ Equation 9

On substitution of these approximations, our expression for the induced birefringence becomes

$$n(\varphi)_e - n_o = n_o\cdot\sqrt{\frac{1}{\cos(\varphi)^2 + \left(1 + 2\cdot\frac{\Delta n}{n}\right)\cdot\sin(\varphi)^2}} - n_o$$

$$n(\varphi)_e - n_o = n_o\cdot\sqrt{\frac{1}{\cos(\varphi)^2 + \left(\sin(\varphi)^2 + 2\cdot\sin(\varphi)^2\cdot\frac{\Delta n}{n}\right)}} - n_o$$

$$n(\varphi)_e - n_o = n_o\cdot\sqrt{\frac{1}{1 + 2\cdot\sin(\varphi)^2\cdot\frac{\Delta n}{n}}} - n_o$$ Equation 10

or approximately

$$n(\varphi)_e - n_o = n_o\cdot\sqrt{1 - 2\cdot\sin(\varphi)^2\cdot\frac{\Delta n}{n}} - n_o$$ Equation 11

$$n(\varphi)_e - n_o = n_o\cdot\left(1 - \sin(\varphi)^2\cdot\frac{\Delta n}{n}\right) - n_o$$ Equation 12

Remembering that

$$\frac{\Delta n}{n} = 1 - \frac{n_e}{n_o}$$ Equation 13

this is

$$n(\varphi)_e - n_o = n_o\cdot\left[1 - \sin(\varphi)^2\cdot\left(1 - \frac{n_e}{n_o}\right)\right] - n_o$$

$$n(\varphi)_e - n_o = n_o - n_o\cdot\sin(\varphi)^2 + \sin(\varphi)^2\cdot n_e - n_o$$

$$n(\varphi)_e - n_o = \sin(\varphi)^2\cdot n_e - n_o\cdot\sin(\varphi)^2$$

$$n(\varphi)_e - n_o = \sin(\varphi)^2 \cdot (n_e - n_o)$$ Equation 14

The angle, φ, is the angle of propagation of the probe beam, measured from the optic axis (which is identified with the pump beam polarisation). The pump beam propagates at the angle, χ, to the Z axis, with polarisation direction inclined from the vertical axis by an angle, κ. The polarisation direction is then given by

$$\text{pump\_polarisation} = \begin{bmatrix} \cos(\kappa) \\ \sin(\kappa)\cdot\cos(\chi) \\ -\sin(\kappa)\cdot\sin(\chi) \end{bmatrix}$$ Equation 15

The cosine of the angle, φ, is obtained from the dot product of the unit probe beam polarisation direction vector and the unit pump beam polarisation vector.

$$\cos(\varphi) = (\text{probe\_propagation})\cdot(\text{pump\_polarisation}) = \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix} \cdot \begin{bmatrix} \cos(\kappa) \\ \sin(\kappa)\cdot\cos(\chi) \\ -\sin(\kappa)\cdot\sin(\chi) \end{bmatrix}$$ Equation 16

$$\cos(\varphi) = (\text{probe\_propagation})\cdot(\text{pump\_polarisation}) = -\sin(\kappa)\cdot\sin(\chi)$$ Equation 17

so that $\sin(\varphi)^2 = 1 - \cos(\varphi)^2 = 1 - \sin(\kappa)^2.\sin(\chi)^2$ Equation 18

Substitution into equation [14], produces the first order approximation to a small induced birefringence

$$n(\varphi)_e - n_o = \left(1 - \sin(\kappa)^2\cdot\sin(\chi)^2\right)\cdot(n_e - n_o)$$ Equation 19

Case 1: Vertical pump beam polarisation, $\kappa = 0$

$$n(\varphi)_e - n_o = n_e - n_o$$ Equation 20

For a vertical pump beam polarisation (i.e. normal to the pump/probe beam intersection plane), the induced birefringence is independent of the intersection angle of pump and probe beams.

Case 2: Horizontal pump beam polarisation, $\kappa = \pi/2$

$$n(\varphi)_e - n_o = \cos(\chi)^2\cdot(n_e - n_o)$$ Equation 21

For a horizontal pump beam polarisation (i.e. lying in the pump/probe beam intersection plane), the induced birefringence decreases according to the square of the cosine of the angle of intersection of pump and probe beams.

Appendix V: Experimental Equipment Specifications

The experiments reported throughout this thesis involve minor variations on two experimental arrangements. An overview of each experiment is given in each of the experimental chapters of this thesis. However, the detailed specifications of the experimental equipment are included in this appendix for reference and to avoid unnecessary repetition.

Schematics of the major experiments

The two major experiments are

- a PLPS experiment (either in an orthogonal or non-orthogonal pump/probe beam geometry) for either a linearly or circularly polarised pump beam, and
- a combined PLPS/PLIF experiment designed to simultaneously image the same target volume with both the PLPS and the PLIF techniques. This experiment was only implemented for a non-orthogonal pump/probe beam geometry, once again, for either a linearly or circularly polarised pump beam.

Schematic diagrams for orthogonal and non-orthogonal PLPS experiments are shown in Figures 1 and 2. The only change in the experiment to implement simultaneous PLIF and PLPS (Figure 3) in the case of a non-orthogonal pump/probe beam geometry was to position a PLIF ICCD camera normal to the pump sheet plane to collect the PLIF fluorescence. The PLPS and PLIF images are then comparable as they both result from the pumped populations in the pump sheet plane and are collected simultaneously.

Equipment

To avoid repetition, the major components of this and the following PLPS imaging experiments are described below. A schematic diagram of the experimental arrangement is included in each chapter to describe additional elements or variations on the basic experimental geometry.

The major experimental elements are

- the laser and frequency doubling system,
- the imaging and timing system,
- the burner/flame systems, and
- the general optical components.

These elements are described below.

Laser and Frequency Doubling System

The laser system consists of Nd:YAG (Continuum Surelite II) pumped dye laser (Lambda Physik Scanmate) operating with Rhodamine 101 to probe the $A^2\Sigma - X^2\Pi$ (0-0) transitions of OH around 308 nm (see Table 1). The Lambda Physik frequency doubling unit produces the required UV output with estimated linewidth, 0.4 cm^{-1}, and pulselength, 3 ns. The dye laser output has linewidth 0.2 cm^{-1} [a] and pulselength 5 ns. The Nd:YAG output at 532 nm (to pump Rhodamine 101) has linewidth 1 cm^{-1} and FWHM pulselength 4-6 ns [b].

Imaging System

Two Princeton Instruments ICCD-576E (576x384 pixel array) cameras were used to collect the PLPS and PLIF image and single point data. The PLIF ICCD was operated with a f2 UV lens. The PLPS ICCD operated without a lens attachment, relying on external UV lenses in a spatial filter arrangement to collect and image the PLPS signal.

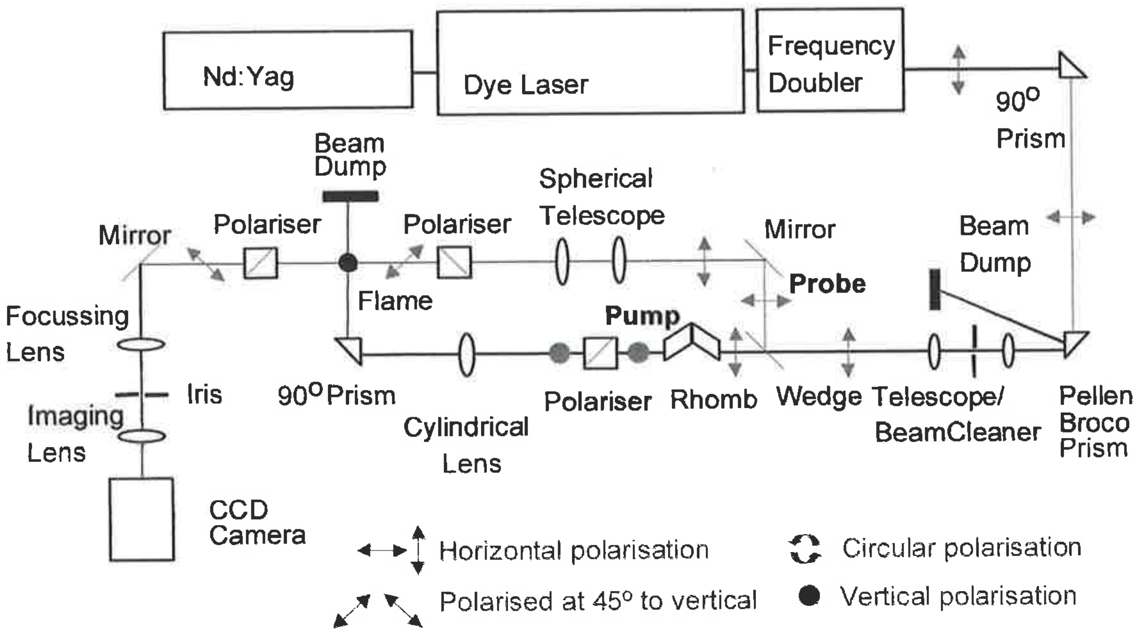

Figure 1: Schematic diagram of the orthogonal PLPS experiment for a vertically polarised pump beam. The laser/doubling system produces a horizontally polarised pump beam. A half-wave rhomb is placed before the pump beam polariser to rotate the plane of polarisation to vertical. The half-wave rhomb is removed in the case of a horizontally polarised pump beam and, in that case, the pump beam polariser is aligned with horizontal transmission axis. In the case of a circularly polarised pump beam, a quarter-wave rhomb is placed in the pump beam path after the polariser and the half-wave rhomb removed. The probe beam is polarised at $\pi/4$ to the vertical.

[a] Lambda Physik Dye Laser Scanmate Instruction Manual, Lambda Physik, 1993.

[b] Continuum Surelite II Manual, Continuum, 1992.

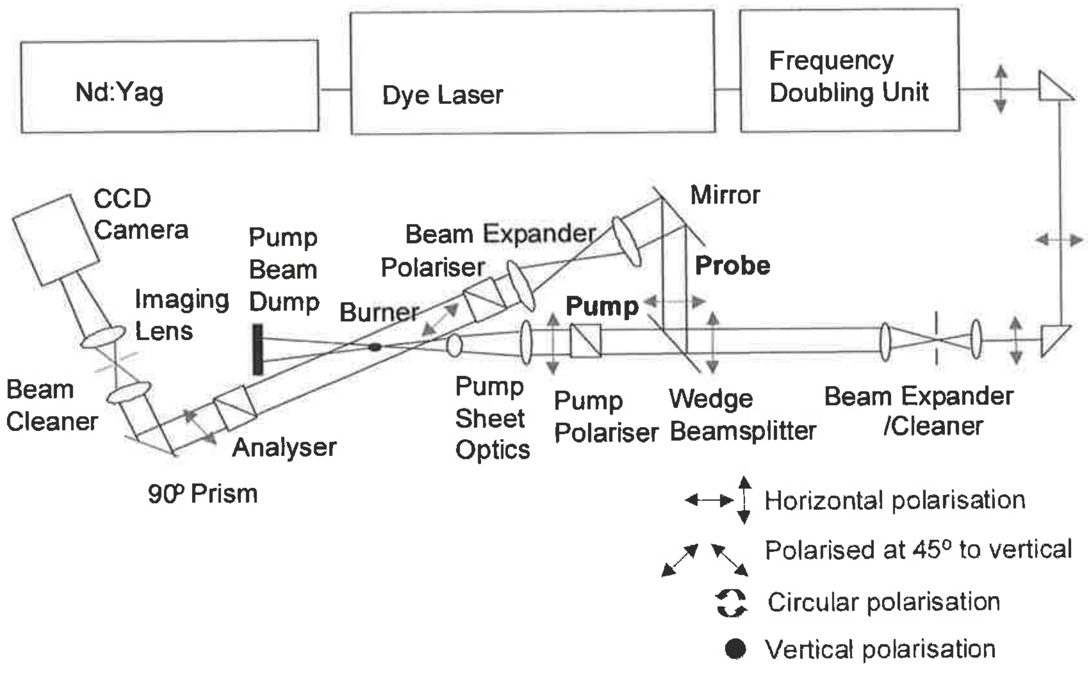

Figure 2: Experimental system for non-collinear PLPS imaging for a horizontally polarised pump beam. The probe beam is polarised at $\pi/4$ to the vertical.

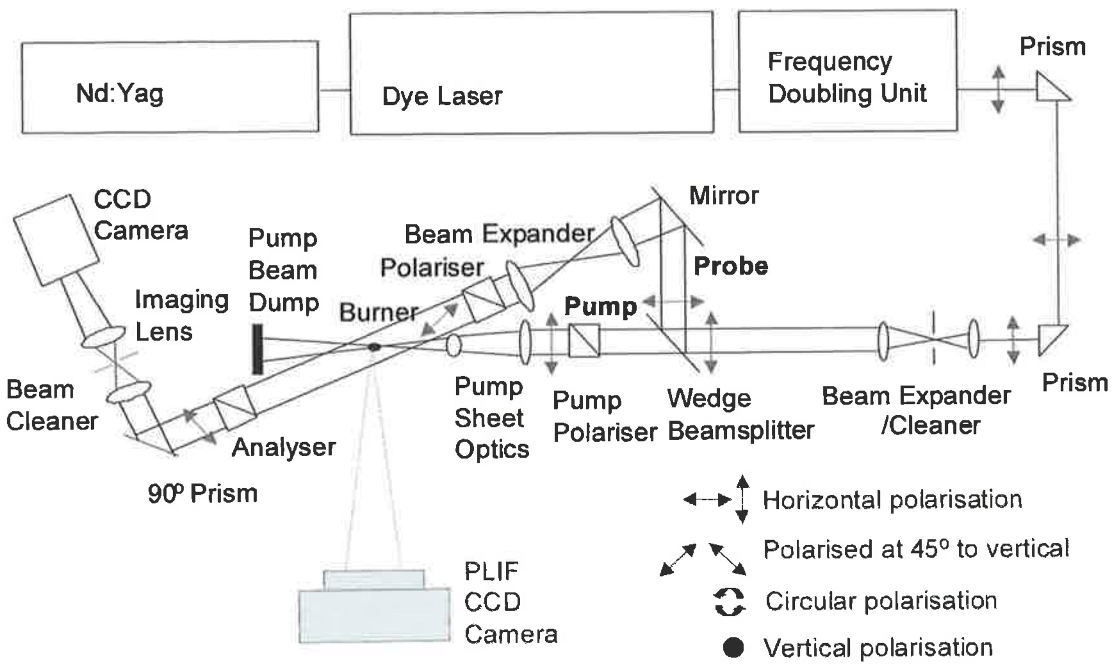

Figure 3: Combined experiment for simultaneous PLIF and PLPS imaging for a horizontally polarised pump beam. The PLIF ICCD is placed normal to the plane of the pump sheet to collect the PLIF fluorescence. The probe beam is polarised at $\pi/4$ to the vertical.

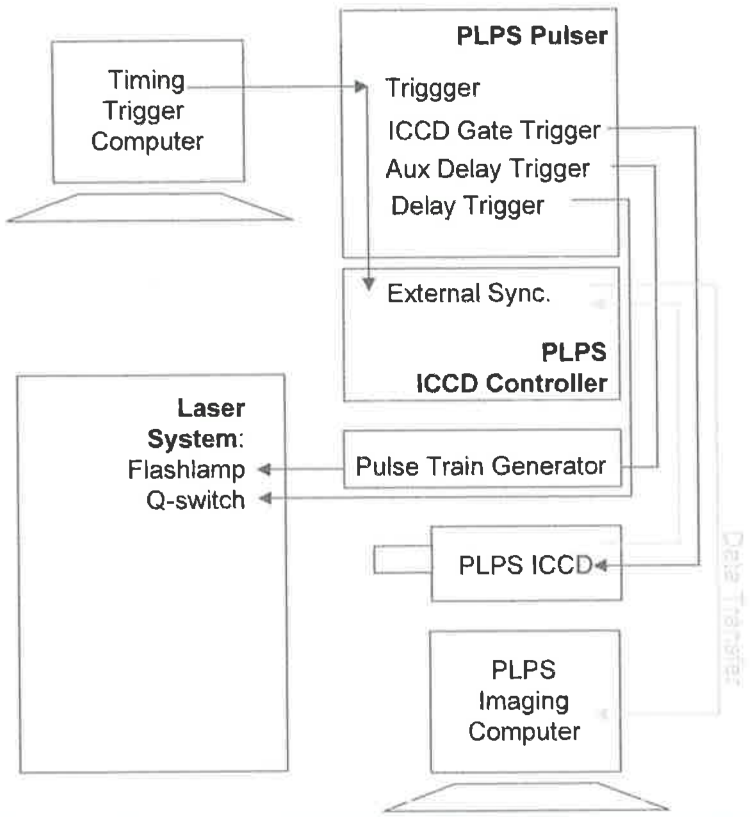

Figure 4: Timing system for single ICCD camera mode operation.

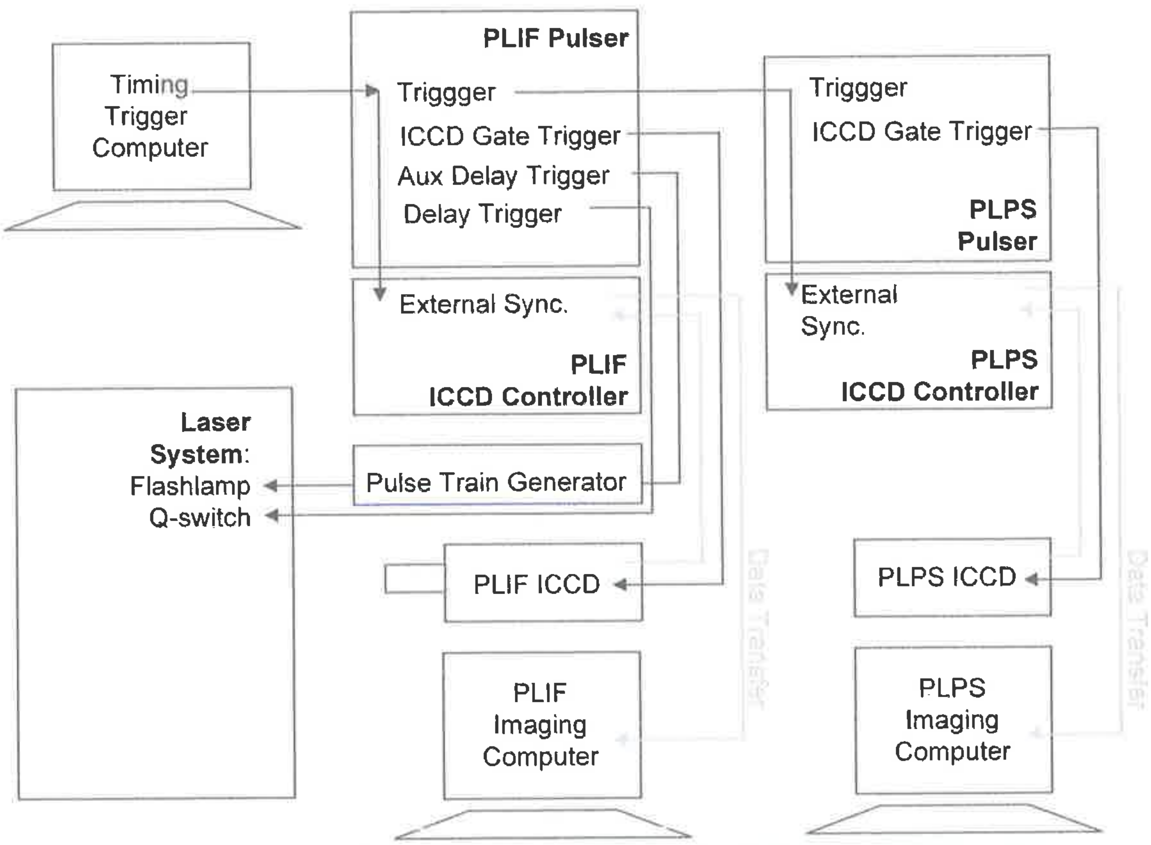

Figure 5: Timing system for simultaneous (dual system) PLPS and PLIF imaging.

Timing System

The timing of the experiment was controlled by a 1 Hz computer generated trigger based on a Pascal program written by Greg Newbold. The one second delay between successive pulses was required to allow downloading of each ICCD image. The system was operated in single or dual camera mode to allow simultaneous PLPS and PLIF imaging. The timing systems for single or dual mode are shown in Figures 4 and 5 respectively. The input trigger was applied in series to two ICCD pulser systems or, for single ICCD mode, to a one ICCD pulser. The pulser, in turn, triggered the ICCD gate with nanosecond resolution to allow imaging with a minimum gating times of 5 ns. The primary pulser in the series additionally sent delayed triggers to the flashlamp and Q-switch of the Nd:YAG laser, synchronised with respect to the gate trigger, to allow the ICCDs to capture the fluorescence (PLIF ICCD) and transmitted probe beam pulse (PLPS ICCD). A control program written by David Johnston allowed remote scanning of the Scanmate dye laser synced to the laser/ICCD system trigger signal. To allow temperature stabilisation of the Nd:YAG laser, a supplementary pulse train generator designed and built by Derek Franklin was connected between the delayed trigger output of the primary pulser and the flashlamp trigger. The pulse train generator triggered the flashlamp at 0.1 second intervals by generating nine equally spaced pulses between each delayed trigger input. The flashlamp was thus maintained at the optimum pulsing rate, 10 Hz, although the Q-switch was only triggered once a second by the imaging pulse.

Burner/Flame Systems for Imaging Experiments

The combustion experiments imaged the OH radical distribution in premixed natural gas/O_2 welding torch type flames. The fuel oxidiser mixture for the imaging experiments was chosen to maximise LPS signal and hence, it is assumed, the concentration of OH. A small modified glass-blowing torch producing a purely premixed flame was used to create flame structures on the scale of the polariser dimensions. The tip of the glass-blowing torch is shown in Figure 6. The tip of the torch has outer diameter 6.2 mm and the main exit orifice has diameter 1.2 mm ± 0.1 mm. There is a secondary circle of small gas orifices surrounding the central 1.2 mm orifice. The modifications to the torch involved removing the standard fuel/oxidiser controls on the handle of the torch and attaching the nozzle to the output of a externally controlled premixed natural gas/O_2 supply. For the OH PLPS flame images in this thesis, the *fuel lean*, *laminar* flame had a Reynolds number estimated to be ~ 95. The flowrates[A2] of the fuel, oxidiser and nitrogen lines were monitored via Fischer and Porter 1/2" and 1/4" flowmeters. Pressure gauges were attached to each flowmeter to determine the operating pressure. The temperature of the fuel flow was assumed to be ambient. For safety, a flashback arrester was attached to the burner inlet port of either the fuel line or the premixed fuel/oxidiser line. Safety blowoff values were also attached to the high pressure cylinders to prevent the line pressure from exceeding safe operation levels for the glass flowmeter tubes .

Figure 6: Tip of the modified glass-blowing burner used in the imaging experiments.

K	P_1	P_2	Q_1	Q_2	R_1	R_2
1	308.2557	-	307.9332	309.1367	307.2899	308.4943
2	308.7283	309.7245	308.0843	309.0756	307.1208	308.1123
3	309.208	310.049	308.2434	309.0756	306.9593	307.7919
4	309.7019	310.424	308.4171	309.1258	306.8129	307.526
5	310.2126	310.8452	308.6089	309.2255	306.6864	307.3089
6	310.744	311.3081	308.8232	309.3681	306.5838	307.1367
7	311.2982	311.8093	309.0629	309.5513	306.5077	307.0066
8	311.8788	312.3469	309.3288	309.7727	306.4613	306.9166
9	312.4848	312.9186	309.6227	310.0308	306.4453	306.8664
10	313.118	313.5245	309.9483	310.3255	306.4613	306.855

Table 1: Wavelengths (in nm) of the $A^2\Sigma - X^2\Pi$ (0-0) transitions of the hydroxyl radical near 310 nm obtained from Dieke and Crosswhite[26].

Gas Supplies

The oxygen (Industrial grade, 020G) was supplied by BOC gases. The natural gas for the glass blowing torch was supplied by high pressure compressed natural gas cylinders (Boral gas).

General Optical Components

The most important optical components for polarisation spectroscopy imaging are the pump/probe beam beamsplitter in the case of a single laser system, and the pump and probe beam polarisers due to the dependence of the signal to background ratio on the polariser extinction ratio.

Glan-Taylor calcite polarisers (Karl Lambrecht) were used for the combustion experiments. An A grade polariser (quoted extinction ratio < $5\text{x}10^{-5}$) was used for the pump beam. Two E grade polarisers were crossed in the probe beam path (quoted extinction ratio < $5\text{x}10^{-6}$). The clear aperture of the polarisers was 20 mm.

The final polarisation component, a fused silica double Fresnel rhomb half wave redarder (Halbo) with 10 mm aperture, was used in either double (half wave) or single (quarter wave) form to rotate the pump plane of polarisation by $^{\pi}/_{2}$ or produce a circularly polarised pump beam.

The quality of PLPS images is directly proportional to the quality of the probe beam profile. Typically a small fraction (<~ 4-10%) of the pump beam is split to form the probe beam in a single laser system. This suggests using the reflection from the front face of a fused silica wedge to form the probe beam. Two options for minimisation of interference fringes on the probe profile due to reflections from front and back faces of the beamsplitter are to

- anti-reflection coat the back face of the beamsplitter plate, and to
- fully separate the front and back face reflections before selecting one component as the probe beam by using a wedge beamsplitter.

An ideal solution would be to use antireflection coating on the back face of a wedge beamsplitter unless it is desired to use the back face reflection to monitor pulse-to-pulse energy. The beamsplitters used in these experiments were 1" fused silica wedges (Casix) designed to deflect the transmitted beam by 6° at 308 nm. No anti-reflection coating was used for the beamsplitting wedges.

The additional optics shown in Figure 1 comprise fused silica lenses and prisms and uncoated UV aluminium mirrors. Unless stated, the diameter of the lenses should be taken to be 1". One CaF_2 lens was used as part of the imaging optics between the probe beam polariser and the imaging ICCD.

The output of the laser/frequency doubling system was passed through a x4 beam expander/cleaner (f_1 = 50 mm, f_2 = 200 mm, pinhole diameter = 200 μm) and split into pump and probe beams by the fused silica wedge. The pump beam polariser and either a $^1/_4$ or $^1/_2$ wave rhomb was used to control the pump beam polarisation. A cylindrical lens (f_3 = 340 mm) was placed with focus at the burner orifice (or for the flat flame burner at the intersection of pump and probe beams within the flame) to produce the pump laser sheet. Measurements of the polarisation state of the pump beam before and after the cylindrical lens indicated that the lens produced negligible change in pump beam polarisation state.

The probe beam was reflected by a UV aluminium mirror and passed through a further X2 telescope (f_4 = 75 mm, f_5 = 150 mm (2")) before transmission through the primary probe beam polariser. The analyser was placed on the far side of the burner. A high resolution polariser rotator (Oriel) allowed rotation of the analyser to ± 0.003° or 5.2 x 10^{-5} radians. An inhouse designed (Jason Peak) Allen key adjustment controlled the vertical and horizontal inclination of the analyser to the probe beam direction.

A second, rectangular, aluminium mirror reflected the transmitted probe beam component towards the PLPS ICCD camera, which was operated without a lens. The beam passed through a focussing lens (f_6 = 200 mm (2")) and an iris passed at the focal plane to minimise scattering from pump and probe beams from reaching the ICCD. A imaging lens (CaF_2 f_7 = 25 mm) in an XY translation mount allowed the flame image to be finely positioned with respect to the CCD array.

References:

[24] Dieke, G.H. and Crosswhite, H.M., *"The Ultraviolet Bands of OH"*, J. Quant. Spectrosc. Radiat. Transfer, **2**, 97-199, 1962.

[26] Conversion calculation methods for the 1/2" and 1/4" flow rates were found in the *"Variable Area Flowmeter Handbook. Vol. 1, Basic Rotameter Principles"*, Fischer and Porter, Catalogue 10A1021 and *"Handbook. Tri-flat Variable-Area Flowmeters, Low flow rate indicators, Data on sizing and calibration prediction"*, Fischer and Porter, Handbook 10A9010 respectively.

Appendix VI: First Order Approximation to the Geometric Dependence of the Induced Circular Birefringence and Optical Activity

Consider the uniaxial gas in the diagonalised geometry described in Appendix II with the addition of optically active behaviour along the optic axis. The displacement vector is related to the electric vector via

$$D=\varepsilon\cdot E \qquad \text{Equation 1}$$

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \begin{bmatrix} \varepsilon_x\cdot E_x \\ \varepsilon_y\cdot E_y \\ \varepsilon_z\cdot E_y \end{bmatrix} + i\cdot\left[\begin{bmatrix} \delta_x \\ \delta_y \\ \delta_z \end{bmatrix} \times \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}\right] \qquad \text{Equation 2}$$

We set the optical activity δ vector to

$$\begin{bmatrix} \delta_x \\ \delta_y \\ \delta_z \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ \delta \end{bmatrix} \qquad \text{Equation 3}$$

and (for the uniaxial material)

$$\varepsilon_x=\varepsilon_y=\varepsilon_o \qquad \text{Equation 4}$$

$$\varepsilon_z=\varepsilon_e \qquad \text{Equation 5}$$

so that

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \begin{bmatrix} \varepsilon_o\cdot E_x \\ \varepsilon_o\cdot E_y \\ \varepsilon_e\cdot E_y \end{bmatrix} + i\cdot\left[\begin{bmatrix} 0 \\ 0 \\ \delta \end{bmatrix} \times \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}\right] = \begin{bmatrix} \varepsilon_o\cdot E_x - i\cdot\delta\cdot E_y \\ \varepsilon_o\cdot E_y + i\cdot\delta\cdot E_x \\ \varepsilon_e\cdot E_y \end{bmatrix} = \begin{bmatrix} \varepsilon_o & -i\cdot\delta & 0 \\ i\cdot\delta & \varepsilon_o & 0 \\ 0 & 0 & \varepsilon_e \end{bmatrix} \cdot \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \qquad \text{Equation 6}$$

Plane wave solution of Maxwell's equations requires that the following equation be satisfied.

$$k^2\cdot E - k\cdot(k\cdot E) - (k_0)^2\cdot D=0 \qquad \text{Equation 7}$$

We choose the principal section as the XZ plane so that

$$k^2=(k_x)^2+(k_z)^2 \quad \text{as} \quad k_y=0 \qquad \text{Equation 8}$$

If the direction of propagation is at the angle φ from the optical axis, we can write

$$k_x=k\cdot\sin(\varphi) \qquad \text{Equation 9}$$

$$k_y=0 \qquad \text{Equation 10}$$

$$k_z = k\cdot\cos(\varphi)$$ Equation 11

and the defining equation becomes

$$\left(k^2\right)\cdot\begin{bmatrix}E_x\\E_y\\E_z\end{bmatrix} - \left[\begin{bmatrix}k\cdot\sin(\varphi)\\0\\k\cdot\cos(\varphi)\end{bmatrix}\cdot\left[\begin{bmatrix}k\cdot\sin(\varphi)\\0\\k\cdot\cos(\varphi)\end{bmatrix}\cdot\begin{bmatrix}E_x\\E_y\\E_z\end{bmatrix}\right]\right] - \left(k_0\right)^2\cdot\left[\begin{bmatrix}\varepsilon_o & -i\cdot\delta & 0\\ i\cdot\delta & \varepsilon_o & 0\\ 0 & 0 & \varepsilon_e\end{bmatrix}\cdot\begin{bmatrix}E_x\\E_y\\E_z\end{bmatrix}\right] = 0$$

Equation 12

or

$$\begin{bmatrix}\left[k^2\cdot\cos(\varphi)^2 - \left(k_0\right)^2\cdot\varepsilon_o\right]\cdot E_x - k^2\cdot\sin(\varphi)\cdot\cos(\varphi)\cdot E_z + i\cdot\left(k_0\right)^2\cdot\delta\cdot E_y\\ \left[k^2 - \left(k_0\right)^2\cdot\varepsilon_o\right]\cdot E_y - i\cdot\left(k_0\right)^2\cdot\delta\cdot E_x\\ \left[k^2 - k^2\cdot\cos(\varphi)^2 - \varepsilon_e\cdot\left(k_0\right)^2\right]\cdot E_z - k^2\cdot\cos(\varphi)\cdot\sin(\varphi)\cdot E_x\end{bmatrix} = 0$$ Equation 13

Dividing through by k_0^2 and replacing the ε factors by their refractive index equivalents gives the matrix equation

$$\begin{bmatrix}n^2\cdot\cos(\varphi)^2 - \left(n_o\right)^2 & i\cdot\delta & -n^2\cdot\sin(\varphi)\cdot\cos(\varphi)\\ -i\cdot\delta & n^2 - \left(n_o\right)^2 & 0\\ -n^2\cdot\sin(\varphi)\cdot\cos(\varphi) & 0 & n^2\cdot\sin(\varphi)^2 - \left(n_e\right)^2\end{bmatrix}\cdot\begin{bmatrix}E_x\\E_y\\E_z\end{bmatrix} = 0$$ Equation 14

Setting the determinant of the left-hand matrix to zero to find the non-trivial solutions produces the condition

$$-\left[\cos(\varphi)^2\cdot\left(n_e\right)^2 + \left(n_o\right)^2\cdot\sin(\varphi)^2\right]\cdot n^4 + \left[\delta^2 - \left(n_o\right)^4\right]\cdot\left(n_e\right)^2 \ldots = 0$$
$$+\left[-\left[\delta^2 - \left(n_o\right)^4\right]\cdot\sin(\varphi)^2 + \left(1+\cos(\varphi)^2\right)\cdot\left(n_o\right)^2\cdot\left(n_e\right)^2\right]\cdot n^2$$ Equation 15

or

$$a\cdot n^4 + b\cdot n^2 + c = 0$$ Equation 16

where

$$a = -\left[\cos(\varphi)^2\cdot\left(n_e\right)^2 + \left(n_o\right)^2\cdot\sin(\varphi)^2\right]$$ Equation 17

$$b = \left[-\left[\delta^2 - \left(n_o\right)^4\right]\cdot\sin(\varphi)^2 + \left(1+\cos(\varphi)^2\right)\cdot\left(n_o\right)^2\cdot\left(n_e\right)^2\right]$$ Equation 18

$$c = \left[\delta^2 - \left(n_o\right)^4\right]\cdot\left(n_e\right)^2$$ Equation 19

Defining

$$\left(n_e\right)^2 = (1+\Delta)\cdot\left(n_o\right)^2$$ Equation 20

and

$$\delta = \sigma \cdot (n_o)^2$$ Equation 21

equations [17] to [19] become

$$a = -(n_o)^2 \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)$$ Equation 22

$$b = (n_o)^4 \cdot 2 \cdot \left[1 + \Delta \cdot \left(\frac{1 + \cos(\varphi)^2}{2}\right) - \frac{\sin(\varphi)^2}{2} \cdot \sigma^2\right]$$ Equation 23

$$c = (n_o)^6 \cdot \left(\sigma^2 - 1\right) \cdot (1 + \Delta)$$ Equation 24

Noting that the term, $\frac{-b}{2 \cdot a}$, may be simplified via

$$\frac{-b}{2 \cdot a} = \frac{2 \cdot \left[1 + \Delta \cdot \left(\frac{1 + \cos(\varphi)^2}{2}\right) - \frac{\sin(\varphi)^2}{2} \cdot \sigma^2\right]}{2 \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)} \cdot (n_o)^2$$

$$\frac{-b}{2 \cdot a} = \frac{\left[2 \cdot \left(1 + \cos(\varphi)^2 \cdot \Delta\right) + \Delta - \cos(\varphi)^2 \cdot \Delta - \sigma^2 + \sigma^2 \cdot \cos(\varphi)^2\right]}{2 \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)} \cdot (n_o)^2$$

$$\frac{-b}{2 \cdot a} = \left[1 + \frac{\left(\Delta - \sigma^2\right) \cdot \sin(\varphi)^2}{2 \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)}\right] \cdot (n_o)^2$$ Equation 25

the two solutions may be rewritten as

$$\frac{(n_\alpha)^2}{(n_o)^2} = \left[1 + \frac{\left(\Delta - \sigma^2\right) \cdot \sin(\varphi)^2}{2 \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)}\right] \cdot \left[1 + \sqrt{1 + \frac{\left(\sigma^2 - 1\right) \cdot (1 + \Delta) \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)}{\left[1 + \Delta \cdot \left(\frac{1 + \cos(\varphi)^2}{2}\right) - \frac{\sin(\varphi)^2}{2} \cdot \sigma^2\right]^2}}\right]$$

Equation 26

and

$$\frac{(n_\beta)^2}{(n_o)^2} = \left[1 + \frac{\left(\Delta - \sigma^2\right) \cdot \sin(\varphi)^2}{2 \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)}\right] \cdot \left[1 - \sqrt{1 + \frac{\left(\sigma^2 - 1\right) \cdot (1 + \Delta) \cdot \left(1 + \Delta \cdot \cos(\varphi)^2\right)}{\left[1 + \Delta \cdot \left(\frac{1 + \cos(\varphi)^2}{2}\right) - \frac{\sin(\varphi)^2}{2} \cdot \sigma^2\right]^2}}\right]$$

Equation 27

Consider two limiting cases of the refractive index solutions.

Case 1: $\varphi = 0$.

$$\frac{(n_\alpha)^2}{(n_o)^2} = (1+0)\cdot\left[1+\sqrt{1+(\sigma^2-1)}\right] = 1+\sigma \qquad \text{Equation 28}$$

and

$$\frac{(n_\beta)^2}{(n_o)^2} = (1+0)\cdot\left[1-\sqrt{1+(\sigma^2-1)}\right] = 1-\sigma \qquad \text{Equation 29}$$

Case 2: $\varphi = \pi/2$.

$$\frac{(n_\alpha)^2}{(n_o)^2} = \left[1+\frac{(\Delta-\sigma^2)}{2}\right]\cdot\left[1+\frac{(\Delta+\sigma^2)}{(2+\Delta-\sigma^2)}\right] = 1+\Delta \qquad \text{Equation 30}$$

and

$$\frac{(n_\beta)^2}{(n_o)^2} = \left[1+\frac{(\Delta-\sigma^2)}{2}\right]\cdot\left[1-\frac{(\Delta+\sigma^2)}{(2+\Delta-\sigma^2)}\right] = 1-\sigma^2 \qquad \text{Equation 31}$$

Any approximation must then take the second term to first order in Δ and σ^2, and the term under the square root sign to second order in Δ and σ^2.

The second term, m,

$$m = \frac{(\Delta-\sigma^2)\cdot\sin(\varphi)^2}{2\cdot(1+\Delta\cdot\cos(\varphi)^2)} \qquad \text{Equation 32}$$

to first order in Δ and σ^2 is

$$m = \left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2 \qquad \text{Equation 33}$$

while the square root term, r,

$$r = \sqrt{1+\frac{(\sigma^2-1)\cdot(1+\Delta)\cdot(1+\Delta\cdot\cos(\varphi)^2)}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]^2}} \qquad \text{Equation 34}$$

is approximated below

$$r=\frac{\sqrt{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]^2+\left(\sigma^2-1\right)\cdot(1+\Delta)\cdot\left(1+\Delta\cdot\cos(\varphi)^2\right)}}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]}$$

$$r=\frac{\sqrt{\begin{array}{l}\left(\frac{1}{4}-\frac{1}{2}\cdot\cos(\varphi)^2+\frac{1}{4}\cdot\cos(\varphi)^4+\sigma^2\cdot\cos(\varphi)^2\right)\cdot\Delta^2\ \ldots\\+\left(\frac{1}{2}\cdot\sigma^2\cdot\cos(\varphi)^4+\frac{1}{2}\cdot\sigma^2+\sigma^2\cdot\cos(\varphi)^2\right)\cdot\Delta\ \ldots\\+\sigma^2\cdot\cos(\varphi)^2+\frac{1}{4}\cdot\sigma^4\cdot\cos(\varphi)^4+\frac{1}{4}\cdot\sigma^4-\frac{1}{2}\cdot\sigma^4\cdot\cos(\varphi)^2\end{array}}}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]}$$

Equation 35

Taking only terms to second order in Δ and σ^2 under the square root sign, this becomes

$$r=\frac{\sqrt{\begin{array}{l}\left(1-2\cdot\cos(\varphi)^2+\cos(\varphi)^4\right)\cdot\frac{\Delta^2}{4}\ \ldots\\+\left(\cos(\varphi)^4+1+2\cdot\cos(\varphi)^2\right)\cdot\frac{\sigma^2}{2}\cdot\Delta\ \ldots\\+\sigma^2\cdot\cos(\varphi)^2+\left(\cos(\varphi)^4+1-2\cdot\cos(\varphi)^2\right)\cdot\frac{\sigma^4}{4}\end{array}}}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]}$$

Equation 36

$$r=\frac{\sqrt{\begin{array}{l}\left(1-2\cdot\cos(\varphi)^2+\cos(\varphi)^4\right)\cdot\frac{\Delta^2}{4}\ \ldots\\+\left(\cos(\varphi)^4+1-2\cdot\cos(\varphi)^2\right)\cdot\frac{\sigma^2}{2}\cdot\Delta+4\cdot\cos(\varphi)^2\cdot\frac{\sigma^2}{2}\cdot\Delta\ \ldots\\+\sigma^2\cdot\cos(\varphi)^2+\left(\cos(\varphi)^4+1-2\cdot\cos(\varphi)^2\right)\cdot\frac{\sigma^4}{4}\end{array}}}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]}$$

$$r=\frac{\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta^2}{4}+\frac{\sigma^2}{2}\cdot\Delta+\frac{\sigma^4}{4}\right)+\cos(\varphi)^2\cdot\left[(2\cdot\Delta+1)\cdot\sigma^2\right]}}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]}$$

$$r=\frac{\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\left[(2\cdot\Delta+1)\cdot\sigma^2\right]}}{\left[1+\Delta\cdot\left(\frac{1+\cos(\varphi)^2}{2}\right)-\frac{\sin(\varphi)^2}{2}\cdot\sigma^2\right]}$$ Equation 37

There are two regions where we can approximate this expression.

Case 1: $\varphi \sim 0$, assuming $\Delta << 1$

$$r_{approx\_1}=\cos(\varphi)\cdot\sigma$$ Equation 38

noting that the denominator is close to unity, while the numerator is of order, σ, allowing us to ignore the Δ term in the denominator.

Case 2: $\varphi \sim \pi/2$

$$r_{approx\_2}=\sin(\varphi)^2\cdot\left(\frac{\Delta+\sigma^2}{2}\right)$$ Equation 39

where we use the same argument to disregard the denominator.

Remembering that

$$m=\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2$$ Equation 40

the approximate solutions in the two cases become

Case 1: $\varphi \sim 0$

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{approx\_1}=\left[1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2\right]\cdot(1+\cos(\varphi)\cdot\sigma)$$ Equation 41

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{approx\_1}=\left[1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2\right]\cdot(1-\cos(\varphi)\cdot\sigma)$$ Equation 42

and to lowest order

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{0\_approx\_1}=1+\cos(\varphi)\cdot\sigma$$ Equation 43

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{0\_approx\_1} = 1 - \cos(\varphi)\cdot\sigma$$ Equation 44

with approximate birefringence, defined as $n_\alpha - n_\beta$, given by

$$\left(\frac{n_\alpha - n_\beta}{n_o}\right)_{0\_approx\_1} = \sqrt{1+\cos(\varphi)\cdot\sigma} \ldots + -\sqrt{1-\cos(\varphi)\cdot\sigma} = \left(1+\frac{\cos(\varphi)\cdot\sigma}{2}\right) - \left(1-\frac{\cos(\varphi)\cdot\sigma}{2}\right) = \cos(\varphi)\cdot\sigma$$

Equation 45

Case 2: $\varphi \sim \pi/2$

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{approx\_2} = \left[1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2\right]\cdot\left[1+\sin(\varphi)^2\cdot\left(\frac{\Delta+\sigma^2}{2}\right)\right]$$ Equation 46

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{approx\_2} = \left[1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2\right]\cdot\left[1-\sin(\varphi)^2\cdot\left(\frac{\Delta+\sigma^2}{2}\right)\right]$$ Equation 47

and to lowest order

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{0\_approx\_2} = 1+\left(\frac{\Delta-\sigma^2}{2}+\frac{\Delta+\sigma^2}{2}\right)\cdot\sin(\varphi)^2 = 1+\Delta\cdot\sin(\varphi)^2$$ Equation 48

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{0\_approx\_2} = 1+\left(\frac{\Delta-\sigma^2}{2}-\frac{\Delta+\sigma^2}{2}\right)\cdot\sin(\varphi)^2 = 1-\sigma^2\cdot\sin(\varphi)^2$$ Equation 49

with approximate birefringence given by

$$\left(\frac{n_\alpha - n_\beta}{n_o}\right)_{0\_approx\_2} = \sqrt{1+\Delta\cdot\sin(\varphi)^2} \ldots + -\sqrt{1-\sigma^2\cdot\sin(\varphi)^2} = \left(1+\frac{\Delta\cdot\sin(\varphi)^2}{2}\right) - \left(1-\frac{\sigma^2\cdot\sin(\varphi)^2}{2}\right)$$

$$\left(\frac{n_\alpha - n_\beta}{n_o}\right)_{0\_approx\_2} = \left(\frac{\Delta+\sigma^2}{2}\right)\cdot\sin(\varphi)^2$$ Equation 50

The nature of the approximations to lowest order suggest we may look again at the full expression for r and hence the general refractive index solution.

Taking only the lowest order terms in equation [37], the general approximation for r becomes

$$r_{approx} = \sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2 + \cos(\varphi)^2\cdot\sigma^2}$$ Equation 51

with general approximate refractive index solutions

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{approx} = \left[1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2\right]\cdot\left[1+\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}\right]$$

Equation 52

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{approx} = \left[1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2\right]\cdot\left[1-\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}\right]$$ Equation 53

and to lowest order

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{0\_approx} = 1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2+\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 54

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{0\_approx} = 1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2-\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 55

with approximate birefringence, n_α - n_β, given by

$$\left(\frac{n_\alpha-n_\beta}{n_o}\right)_{0\_approx} = \sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 56

Note that the birefringence for propagation along the optic axis is due to the optical activity, while the birefringence for propagation normal to the optic axis has components due to both the linear birefringence and the optical activity, although the optically active component occurs to second order only in this case.

Note also that for a non-optically active medium, $\delta = \sigma = 0$, the expression for the induced birefringence reduces to

$$\left(\frac{n_\alpha-n_\beta}{n_o}\right)_{0\_approx\ ...\ +non-optically\_active} = \sin(\varphi)^2\cdot\frac{\Delta}{2}$$ Equation 57

with maximum induced birefringence for probe propagation normal to the induced optic axis and zero induced birefringence for propagation parallel to the induced optic axis.

Calculation of the Electric Field Components

The electric field components may be calculated from equation [14]

$$\begin{bmatrix} n^2\cdot\cos(\varphi)^2-(n_o)^2 & i\cdot\delta & -n^2\cdot\sin(\varphi)\cdot\cos(\varphi) \\ -i\cdot\delta & n^2-(n_o)^2 & 0 \\ -n^2\cdot\sin(\varphi)\cdot\cos(\varphi) & 0 & n^2\cdot\sin(\varphi)^2-(n_e)^2 \end{bmatrix}\cdot\begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}=0 \qquad \text{Equation 58}$$

which may be expanded to give

$$\begin{bmatrix} \left[n^2\cdot\cos(\varphi)^2-(n_o)^2\right]\cdot E_x+i\cdot\delta\cdot E_y-n^2\cdot\sin(\varphi)\cdot\cos(\varphi)\cdot E_z \\ -i\cdot\delta\cdot E_x+\left[n^2-(n_o)^2\right]\cdot E_y \\ \left[n^2\cdot\sin(\varphi)^2-(n_e)^2\right]\cdot E_z-n^2\cdot\sin(\varphi)\cdot\cos(\varphi)\cdot E_x \end{bmatrix}=0 \qquad \text{Equation 59}$$

The second component gives the relationship

$$E_y=i\cdot\left[\frac{\delta}{n^2-(n_o)^2}\right]\cdot E_x \qquad \text{Equation 60}$$

and the third

$$E_z=\left[\frac{n^2\cdot\sin(\varphi)\cdot\cos(\varphi)}{n^2\cdot\sin(\varphi)^2-(n_e)^2}\right]\cdot E_x=\left[\frac{n^2\cdot\sin(\varphi)\cdot\cos(\varphi)}{n^2\cdot\sin(\varphi)^2-(1+\Delta)\cdot(n_o)^2}\right]\cdot E_x \qquad \text{Equation 61}$$

Equations [60] and [61] assume that the denominators in each case are non-zero.

The unnormalised electric vector is then given by

$$E=\begin{bmatrix} 1 \\ i\cdot\left[\dfrac{\delta}{n^2-(n_o)^2}\right] \\ \dfrac{n^2\cdot\sin(\varphi)\cdot\cos(\varphi)}{n^2\cdot\sin(\varphi)^2-(1+\Delta)\cdot(n_o)^2} \end{bmatrix} \qquad \text{Equation 62}$$

Noting that we may write

$$\left(\frac{n}{n_o}\right)^2=1+s \qquad \text{Equation 63}$$

where s << 1, the Z component may be approximated by

$$E_{z\_approx} = \frac{\sin(\varphi)\cdot\cos(\varphi)}{\sin(\varphi)^2 - \left(\frac{1+\Delta}{1+s}\right)} = \frac{\sin(\varphi)\cdot\cos(\varphi)}{\sin(\varphi)^2 - (1+\Delta)\cdot(1-s)}$$

$$E_{z\_approx} = \frac{\sin(\varphi)\cdot\cos(\varphi)}{\sin(\varphi)^2 - (1 - s + \Delta - \Delta\cdot s)}$$

and to first order

$$E_{z\_approx} = \frac{\sin(\varphi)\cdot\cos(\varphi)}{-\cos(\varphi)^2 + (s - \Delta + \Delta\cdot s)}$$ Equation 64

<u>For case 1</u>: $\varphi \sim 0$, and $\cos(\varphi) \sim 1$, this is approximately

$$E_{z\_0\_approx\_1} = -\tan(\varphi)$$ Equation 65

however, for case 2: $\varphi \sim \pi/2$ and for the general case, the following expression must be used.

$$E_{z\_0\_approx\_2} = \frac{\sin(\varphi)\cdot\cos(\varphi)}{-\cos(\varphi) + (s - \Delta)}$$ Equation 66

The electric field vector may then be approximated by the vector

$$E_{0\_approx} = \begin{bmatrix} 1 \\ i\cdot\left(\frac{\sigma}{s}\right) \\ \frac{\sin(\varphi)\cdot\cos(\varphi)}{\left[-\cos(\varphi)^2 + (s-\Delta)\right]} \end{bmatrix}$$ Equation 67

or

$$E_{0\_approx} = \begin{bmatrix} \cos(\varphi)^2 - (s-\Delta) \\ i\cdot\left[\frac{\sigma\cdot\left[\cos(\varphi)^2 - (s-\Delta)\right]}{s}\right] \\ -\sin(\varphi)\cdot\cos(\varphi) \end{bmatrix}$$ Equation 68

where the terms $(s - \Delta)$ may be omitted if $\varphi \sim 0$.

To eliminate infinities when the term, s, is zero, we multiply through by s so that

$$E_{0\_approx} = \begin{bmatrix} \left[\cos(\varphi)^2 - (s-\Delta)\right]\cdot s \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - (s-\Delta)\right]\right] \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot s \end{bmatrix}$$ Equation 69

For case 1: $\varphi \sim 0$, the s functions defined in equation [62] are given by

$$s_{\alpha\_0\_approx\_1} = \cos(\varphi)\cdot\sigma$$ Equation 70

$$s_{\beta\_0\_approx\_1} = -\cos(\varphi)\cdot\sigma$$ Equation 71

so that, using the full approximation in the new normalisation of equation [69]

$$E_{\alpha\_0\_approx\_1} = \begin{bmatrix} \left[\cos(\varphi)^2 - (\cos(\varphi)\cdot\sigma - \Delta)\right]\cdot\cos(\varphi)\cdot\sigma \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - (\cos(\varphi)\cdot\sigma - \Delta)\right]\right] \\ -\sin(\varphi)\cdot\cos(\varphi)^2\cdot\sigma \end{bmatrix}$$ Equation 72

which, assuming $\cos(\varphi) >> \sigma, \Delta$, becomes

$$E_{\alpha\_0\_approx\_1} = \begin{bmatrix} \cos(\varphi)^3\cdot\sigma \\ i\cdot\cos(\varphi)^2\cdot\sigma \\ -\sin(\varphi)\cdot\cos(\varphi)^2\cdot\sigma \end{bmatrix} = \sigma\cdot\cos(\varphi)^2\cdot\begin{bmatrix} \cos(\varphi) \\ i \\ -\sin(\varphi) \end{bmatrix}$$ Equation 73

Similarly

$$E_{\beta\_0\_approx\_1} = \begin{bmatrix} \left[\cos(\varphi)^2 - (-\cos(\varphi)\cdot\sigma - \Delta)\right]\cdot(-\cos(\varphi)\cdot\sigma) \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - (-\cos(\varphi)\cdot\sigma - \Delta)\right]\right] \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot(-\cos(\varphi)\cdot\sigma) \end{bmatrix}$$ Equation 74

becomes

$$E_{\beta\_0\_approx\_1} = \begin{bmatrix} -\cos(\varphi)^3\cdot\sigma \\ i\cdot\cos(\varphi)^2\cdot\sigma \\ \sin(\varphi)\cdot\cos(\varphi)^2\cdot\sigma \end{bmatrix} = -\sigma\cdot\cos(\varphi)^2\cdot\begin{bmatrix} \cos(\varphi) \\ -i \\ -\sin(\varphi) \end{bmatrix}$$ Equation 75

so that, in summary, for case 1: $\varphi \sim 0$.

$$E_{\alpha\_0\_approx\_1} = \sigma\cdot\cos(\varphi)^2\cdot\begin{bmatrix} \cos(\varphi) \\ i \\ -\sin(\varphi) \end{bmatrix}$$ Equation 76

and

$$E_{\beta\_0\_approx\_1} = -\sigma\cdot\cos(\varphi)^2\cdot\begin{bmatrix} \cos(\varphi) \\ -i \\ -\sin(\varphi) \end{bmatrix}$$ Equation 77

The vector components of these two expressions represent orthogonal circular polarisation states independent of the optically active constant, σ.

For case 2: $\varphi \sim \pi/2$, the s functions defined in equation [62] are given by

$$s_{\alpha\_0\_approx\_2} = \Delta\cdot\sin(\varphi)^2$$ Equation 78

$$s_{\beta\_0\_approx\_2} = -\sigma^2\cdot\sin(\varphi)^2$$ Equation 79

Remembering that the unnormalised electric field vector is

$$E_{0\_approx} = \begin{bmatrix} \left[\cos(\varphi)^2 - (s - \Delta)\right]\cdot s \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - (s-\Delta)\right]\right] \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot s \end{bmatrix} \qquad \text{Equation 80}$$

the approximate α electric field component is then given by

$$E_{\alpha\_0\_approx\_2} = \begin{bmatrix} \left[\cos(\varphi)^2 - \left(\Delta\cdot\sin(\varphi)^2 - \Delta\right)\right]\cdot\left(\Delta\cdot\sin(\varphi)^2\right) \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - \left(\Delta\cdot\sin(\varphi)^2 - \Delta\right)\right]\right] \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot\left(\Delta\cdot\sin(\varphi)^2\right) \end{bmatrix} \qquad \text{Equation 81}$$

$$E_{\alpha\_0\_approx\_2} = \begin{bmatrix} \left[\cos(\varphi)^2 - \left(-\Delta\cdot\cos(\varphi)^2\right)\right]\cdot\left(\Delta\cdot\sin(\varphi)^2\right) \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - \left(-\Delta\cdot\cos(\varphi)^2\right)\right]\right] \\ -\sin(\varphi)^3\cdot\cos(\varphi)\cdot\Delta \end{bmatrix}$$

$$E_{\alpha\_0\_approx\_2} = \begin{bmatrix} \left[\cos(\varphi)^2\cdot(1+\Delta)\right]\cdot\left(\Delta\cdot\sin(\varphi)^2\right) \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2\cdot(1+\Delta)\right]\right] \\ -\sin(\varphi)^3\cdot\cos(\varphi)\cdot\Delta \end{bmatrix} \qquad \text{Equation 82}$$

which, approximating the $(1 + \Delta)$ terms by unity becomes

$$E_{\alpha\_0\_approx\_2} = \Delta\cdot\cos(\varphi)\cdot\sin(\varphi)^2\cdot\begin{bmatrix} \cos(\varphi) \\ i\cdot\left(\frac{\sigma}{\Delta}\right)\cdot\frac{\cos(\varphi)}{\sin(\varphi)^2} \\ -\sin(\varphi) \end{bmatrix} \qquad \text{Equation 83}$$

Similarly, the β electric field component is given by

$$E_{\beta\_0\_approx\_2} = \begin{bmatrix} \left[\cos(\varphi)^2 - \left(-\sigma^2\cdot\sin(\varphi)^2 - \Delta\right)\right]\cdot\left(-\sigma^2\cdot\sin(\varphi)^2\right) \\ i\cdot\left[\sigma\cdot\left[\cos(\varphi)^2 - \left(-\sigma^2\cdot\sin(\varphi)^2 - \Delta\right)\right]\right] \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot\left(-\sigma^2\cdot\sin(\varphi)^2\right) \end{bmatrix} \qquad \text{Equation 84}$$

or, writing

$$\cos(\varphi)^2 + \sigma^2\cdot\sin(\varphi)^2 + \Delta = \cos(\varphi)^2\cdot\left(1-\sigma^2\right) + \sigma^2 + \Delta \qquad \text{Equation 85}$$

and approximating $1 - \sigma^2$ by unity, so that we may approximate

$$\cos(\varphi)^2 + \sigma^2\cdot\sin(\varphi)^2 + \Delta = \cos(\varphi)^2 + \sigma^2 + \Delta \qquad \text{Equation 86}$$

we have

$$E_{\beta\_0\_approx\_2} = \begin{bmatrix} \left(\cos(\varphi)^2 + \sigma^2 + \Delta\right)\cdot\left(-\sigma^2\cdot\sin(\varphi)^2\right) \\ i\cdot\left[\sigma\cdot\left(\cos(\varphi)^2 + \sigma^2 + \Delta\right)\right] \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot\left(-\sigma^2\cdot\sin(\varphi)^2\right) \end{bmatrix} \qquad \text{Equation 87}$$

or

$$E_{\beta\_0\_approx\_2} = -\sigma^2\cdot\sin(\varphi)^2\cdot\cos(\varphi)\cdot\begin{bmatrix} \dfrac{\cos(\varphi)^2+\sigma^2+\Delta}{\cos(\varphi)} \\ i\cdot\left[\dfrac{1}{\sigma}\cdot\left(\dfrac{\cos(\varphi)^2+\sigma^2+\Delta}{\cos(\varphi)\cdot\sin(\varphi)^2}\right)\right] \\ -\sin(\varphi) \end{bmatrix}$$ Equation 88

An infinity may occur in equation [88] for $\varphi = {}^{\pi}/_{2}$ or $\sigma = 0$, so we rewrite equations [83] and [88] as

$$E_{\alpha\_0\_approx\_2} = \Delta\cdot\sin(\varphi)^2\cdot\begin{bmatrix} \cos(\varphi)^2 \\ i\cdot\left(\dfrac{\sigma}{\Delta}\right)\cdot\dfrac{\cos(\varphi)^2}{\sin(\varphi)^2} \\ -\sin(\varphi)\cdot\cos(\varphi) \end{bmatrix}$$ Equation 89

$$E_{\beta\_0\_approx\_2} = -\sigma\cdot\sin(\varphi)^2\cdot\begin{bmatrix} \left(\cos(\varphi)^2+\sigma^2+\Delta\right)\cdot\sigma \\ i\cdot\left(\dfrac{\cos(\varphi)^2+\sigma^2+\Delta}{\sin(\varphi)^2}\right) \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot\sigma \end{bmatrix}$$ Equation 90

For $\varphi = \pi/2$, the two unnormalised polarisation modes become

$$E_{\alpha\_0\_approx\_2} = \Delta\cdot\sin(\varphi)^2\cdot\begin{bmatrix} 0 \\ 0 \\ -1 \end{bmatrix}\cdot\cos(\varphi)$$ Equation 91

$$E_{\beta\_0\_approx\_2} = -\sigma\cdot\sin(\varphi)^2\cdot\begin{bmatrix} -i\cdot\sigma \\ 1 \\ 0 \end{bmatrix}\cdot i\cdot\left(\sigma^2+\Delta\right)$$ Equation 92

We can see that the α polarisation mode lies along the Z axis of the diagonalised geometry, while the β polarisation mode lies very close to the Y axis, with a small X component due to the induced circular birefringence.

Summarising the results for the two cases and attempting to write them in the same format, we have

<u>Case I</u>: $\varphi \sim 0$

$$E_{\alpha\_0\_approx\_1} = \sigma\cdot\cos(\varphi)\cdot\begin{bmatrix} \cos(\varphi)^2 \\ i\cdot\cos(\varphi) \\ -\sin(\varphi)\cdot\cos(\varphi) \end{bmatrix}$$ Equation 93

and

$$E_{\beta\_0\_approx\_1} = -\sigma\cdot\cos(\varphi)\cdot\begin{bmatrix}\cos(\varphi)^2\\ -i\cdot\cos(\varphi)\\ -\sin(\varphi)\cdot\cos(\varphi)\end{bmatrix}$$ Equation 94

<u>Case 2</u>: $\varphi \sim \pi/2$

$$E_{\alpha\_0\_approx\_2} = \Delta\cdot\sin(\varphi)^2\cdot\begin{bmatrix}\cos(\varphi)^2\\ i\cdot\left(\frac{\sigma}{\Delta}\right)\cdot\frac{\cos(\varphi)^2}{\sin(\varphi)^2}\\ -\sin(\varphi)\cdot\cos(\varphi)\end{bmatrix}$$ Equation 95

$$E_{\beta\_0\_approx\_2} = -\sigma^2\cdot\sin(\varphi)^2\cdot\left[\begin{bmatrix}\cos(\varphi)^2\\ i\cdot\left(\frac{1}{\sigma}\right)\cdot\left(\frac{\cos(\varphi)^2}{\sin(\varphi)^2}\right)\\ -\sin(\varphi)\cdot\cos(\varphi)\end{bmatrix}+\begin{bmatrix}\left(\sigma^2+\Delta\right)\\ i\cdot\left(\frac{\sigma^2+\Delta}{\sigma}\right)\cdot\left(\frac{1}{\sin(\varphi)^2}\right)\\ -\sin(\varphi)\cdot\cos(\varphi)\end{bmatrix}\right]$$ Equation 96

The form of these equations suggest that a general electric field vector may be obtained where $\cos^2(\varphi) >> \sigma^2$, Δ and the polarisation modes lie in the plane of polarisation of the probe beam, i.e. the Z component is proportional to $\sin(\varphi)$ and the X component is proportional to $\cos(\varphi)$.

Remembering that the general refractive index solutions are

$$\left[\left(\frac{n_\alpha}{n_o}\right)^2\right]_{0\_approx} = 1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2+\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 97

and

$$\left[\left(\frac{n_\beta}{n_o}\right)^2\right]_{0\_approx} = 1+\left(\frac{\Delta-\sigma^2}{2}\right)\cdot\sin(\varphi)^2-\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 98

so that

$$s_{\alpha\_0\_approx} = \frac{\Delta-\sigma^2}{2}\cdot\sin(\varphi)^2+\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 99

and

$$s_{\beta\_0\_approx} = \frac{\Delta-\sigma^2}{2}\cdot\sin(\varphi)^2-\sqrt{\sin(\varphi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\varphi)^2\cdot\sigma^2}$$ Equation 100

the general electric field vectors, assuming $\cos^2(\varphi) >> \sigma^2$, Δ (and hence $\cos(\varphi) > 0$) are

$$E_{\alpha\_0\_approx} = \begin{bmatrix} \cos(\varphi)^2 \cdot s_\alpha \\ i \cdot \sigma \cdot \cos(\varphi)^2 \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot s_\alpha \end{bmatrix} = s_\alpha \cdot \begin{bmatrix} \cos(\varphi)^2 \\ i \cdot \frac{\sigma}{s_\alpha} \cdot \cos(\varphi)^2 \\ -\sin(\varphi)\cdot\cos(\varphi) \end{bmatrix}$$ Equation 101

and

$$E_{\beta\_0\_approx} = \begin{bmatrix} \cos(\varphi)^2 \cdot s_\beta \\ i \cdot \sigma \cdot \cos(\varphi)^2 \\ -\sin(\varphi)\cdot\cos(\varphi)\cdot s_\beta \end{bmatrix} = s_\beta \cdot \begin{bmatrix} \cos(\varphi)^2 \\ i \cdot \frac{\sigma}{s_\beta} \cdot \cos(\varphi)^2 \\ -\sin(\varphi)\cdot\cos(\varphi) \end{bmatrix}$$ Equation 102

Note as $\cos(\varphi) > 0$, we have no difficulties with infinities with the s factor in the denominator.

We now move from the above diagonalised axes and assume a variation on the usual polarisation spectroscopy axes in order to apply the α and β induced birefringence to the two probe beam polarisation mode components in an experimental configuration.

Let the pump beam propagate along Z axis with polarisation axis parallel to

$$A_{pump} = \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix}$$ Equation 103

and the probe beam propagate at the angle, χ, to the Z axis on the YZ plane, polarised at an angle, γ, to the vertical X axis. The probe beam propagation direction is

$$k_{probe} = \begin{bmatrix} 0 \\ \sin(\chi) \\ \cos(\chi) \end{bmatrix}$$ Equation 104

and polarisation direction

$$\frac{E_{probe}}{E_{probe\_0}} = \begin{bmatrix} \cos(\gamma) \\ \sin(\gamma)\cdot\cos(\chi) \\ -\sin(\gamma)\cdot\sin(\chi) \end{bmatrix}$$ Equation 105

where E_{probe_0} is the magnitude of the probe beam electric field incident on the primary probe beam polariser.

The angle, χ, is equivalent to the angle, φ, between probe beam propagation direction and the induced optic axis as the polarisation axis of the circularly polarised pump beam lies along its direction of propagation.

The primary probe beam polariser is assumed parallel to the probe beam polarisation

$$A_{probe} = \begin{bmatrix} \cos(\gamma) \\ \sin(\gamma)\cdot\cos(\chi) \\ -\sin(\gamma)\cdot\sin(\chi) \end{bmatrix}$$ Equation 106

with crossed analyser at

$$A_{analyser} = \begin{bmatrix} -\sin(\gamma) \\ \cos(\gamma)\cdot\cos(\chi) \\ -\cos(\gamma)\cdot\sin(\chi) \end{bmatrix}$$ Equation 107

Consider the general case where we assume that $\cos^2(\varphi) >> \sigma^2, \Delta$

We rearrange the general polarisation modes, swapping X and Y components to reflect this geometry. We also ignore the coefficient, s, and divide through by the factor, $\cos(\varphi)$ (or $\cos(\chi)$ as $\varphi = \chi$) as $\cos(\varphi)$ is not-zero for this condition.

The electric field components are then written

$$E_{\alpha\_0\_approx} = \begin{bmatrix} i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi) \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix}$$ Equation 108

and

$$E_{\beta\_0\_approx} = \begin{bmatrix} i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi) \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix}$$ Equation 109

The electric field of the probe beam must be expressed in terms of the α and β electric field vectors. As the vectors are not normalised, we write

$$\frac{E_{probe}}{E_{probe\_0}} = f\cdot E_{\alpha\_0\_approx} + g\cdot E_{\beta\_0\_approx}$$ Equation 110

Let us describe the system with respect to the distance, Λ, travelled along the α and β ray paths. After passage through a distance, Λ, through the dichroism and birefringent region, the probe beam is

$$\frac{E_{probe\_transmitted}}{E_{probe\_0}} = f\cdot E_{\alpha\_0\_approx}\cdot e^{-\frac{\alpha_\alpha}{2}\cdot\Lambda + i\cdot k_\alpha\cdot\Lambda} + g\cdot E_{\beta\_0\_approx}\cdot e^{-\frac{\alpha_\beta}{2}\cdot\Lambda + i\cdot k_\beta\cdot\Lambda}$$ Equation 111

Letting

$$\alpha_\alpha = \alpha + \frac{\Delta\alpha}{2} \qquad k_\alpha = k + \frac{\Delta k}{2}$$ Equation 112

$$\alpha_\beta = \alpha - \frac{\Delta\alpha}{2} \qquad k_\beta = k - \frac{\Delta k}{2}$$ Equation 113

the transmitted probe field is

$$\frac{E_{probe\_transmitted}}{E_{probe\_0}} = \begin{bmatrix} f \cdot E_{\alpha\_0\_approx} \cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \ldots \\ + g \cdot E_{\beta\_0\_approx} \cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda} \end{bmatrix} \cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$ Equation 114

The fraction of the probe beam transmitted through the crossed analyser is then

$$\frac{E_{probe\_transmitted\_analyser}}{E_{probe\_0}} = \begin{bmatrix} f \cdot \left(E_{\alpha\_0\_approx} \cdot A_{analyser}\right) \cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \ldots \\ + g \cdot \left(E_{\beta\_0\_approx} \cdot A_{analyser}\right) \cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda} \end{bmatrix} \cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$ Equation 115

To determine the f and g factors, we rewrite equation [110]

$$\begin{bmatrix} \cos(\gamma) \\ \sin(\gamma)\cdot\cos(\chi) \\ -\sin(\gamma)\cdot\sin(\chi) \end{bmatrix} = f \cdot \begin{bmatrix} i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi) \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix} + g \cdot \begin{bmatrix} i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi) \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix}$$ Equation 116

giving the two simultaneous equations,

$$\begin{bmatrix} i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi) & i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi) \\ 1 & 1 \end{bmatrix} \cdot \begin{bmatrix} f \\ g \end{bmatrix} = \begin{bmatrix} \cos(\gamma) \\ \sin(\gamma) \end{bmatrix}$$ Equation 117

or

$$\begin{bmatrix} f \\ g \end{bmatrix} = \begin{bmatrix} i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi) & i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi) \\ 1 & 1 \end{bmatrix}^{-1} \cdot \begin{bmatrix} \cos(\gamma) \\ \sin(\gamma) \end{bmatrix}$$ Equation 118

$$\begin{bmatrix} f \\ g \end{bmatrix} = \frac{i}{\sigma\cdot\cos(\chi)} \cdot \left(\frac{s_\alpha \cdot s_\beta}{s_\alpha - s_\beta}\right) \cdot \left[\begin{bmatrix} 1 & -i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi) \\ -1 & i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi) \end{bmatrix} \cdot \begin{bmatrix} \cos(\gamma) \\ \sin(\gamma) \end{bmatrix}\right]$$ Equation 119

$$\begin{bmatrix} f \\ g \end{bmatrix} = \frac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)\cdot\begin{bmatrix} -i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi)\cdot\sin(\gamma)+\cos(\gamma) \\ -1\cdot\left(-i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi)\cdot\sin(\gamma)+\cos(\gamma)\right) \end{bmatrix}$$

Equation 120

while

$$E_{\alpha\_0\_approx}\cdot A_{analyser} = \begin{bmatrix} i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi) \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix}\cdot\begin{bmatrix} -\sin(\gamma) \\ \cos(\gamma)\cdot\cos(\chi) \\ -\cos(\gamma)\cdot\sin(\chi) \end{bmatrix}$$

Equation 121

$$E_{\alpha\_0\_approx}\cdot A_{analyser} = -i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi)\cdot\sin(\gamma)+\cos(\gamma)$$

Equation 122

and

$$E_{\beta\_0\_approx}\cdot A_{analyser} = \begin{bmatrix} i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi) \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix}\cdot\begin{bmatrix} -\sin(\gamma) \\ \cos(\gamma)\cdot\cos(\chi) \\ -\cos(\gamma)\cdot\sin(\chi) \end{bmatrix}$$

Equation 123

$$E_{\beta\_0\_approx}\cdot A_{analyser} = -i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi)\cdot\sin(\gamma)+\cos(\gamma)$$

Equation 124

so that

$$\frac{f\cdot\left(E_{\alpha\_0\_approx}\cdot A_{analyser}\right)}{\frac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)} = \left(-i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi)\cdot\sin(\gamma)+\cos(\gamma)\right)\cdot\left(-i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi)\cdot\sin(\gamma)+\cos(\gamma)\right)$$

Equation 125

$$\begin{aligned}\frac{f\cdot\left(E_{\alpha\_0\_approx}\cdot A_{analyser}\right)}{\frac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)} = & -i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi)\cdot\sin(\gamma)\cdot\left(-i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi)\cdot\sin(\gamma)\right)\ \ldots \\ & +\cos(\gamma)\cdot\left(-i\cdot\frac{\sigma}{s_\beta}\cdot\cos(\chi)\cdot\sin(\gamma)+-i\cdot\frac{\sigma}{s_\alpha}\cdot\cos(\chi)\cdot\sin(\gamma)\right)\ \ldots \\ & +\cos(\gamma)^2\end{aligned}$$

Equation 126

$$\begin{aligned}\frac{f\cdot\left(E_{\alpha\_0\_approx}\cdot A_{analyser}\right)}{\frac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)} = -\frac{g\cdot\left(E_{\beta\_0\_approx}\cdot A_{analyser}\right)}{\frac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)} = H = & \cos(\gamma)^2-\left(\frac{\sigma}{s_\beta}\right)\cdot\left(\frac{\sigma}{s_\alpha}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\ \ldots \\ & +-i\cdot\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{s_\beta}+\frac{\sigma}{s_\alpha}\right)\end{aligned}$$

Equation 127

giving the transmitted probe beam electric field

$$\frac{\dfrac{E_{probe\_transmitted\_analyser}}{E_{probe\_0}}}{\dfrac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\dfrac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda}\cdot e^{i\cdot k\cdot\Lambda}} = H\cdot\left(e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \dots + -e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda}\right)$$

Equation 128

$$\frac{\dfrac{E_{probe\_transmitted\_analyser}}{E_{probe\_0}}}{\dfrac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\dfrac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda}\cdot e^{i\cdot k\cdot\Lambda}} = H\cdot\left[e^{i\cdot\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda} - e^{-i\cdot\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda}\right]$$

Equation 129

or

$$\frac{\dfrac{E_{probe\_transmitted\_analyser}}{E_{probe\_0}}}{\dfrac{i}{\sigma\cdot\cos(\chi)}\cdot\left(\dfrac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda}\cdot e^{i\cdot k\cdot\Lambda}} = H\cdot\left[2i\cdot\sin\left[\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]\right]$$

Equation 130

The transmitted probe beam intensity is then

$$\frac{\dfrac{I_{probe\_transmitted\_analyser}}{I_{probe\_0}}}{\dfrac{1}{(\sigma\cdot\cos(\chi))^2}\cdot\left(\dfrac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)^2\cdot e^{-\alpha\cdot\Lambda}} = 4\cdot\left[\sin\left[\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]\cdot\sin\left[\left(\frac{\Delta k}{2}-i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]\right]\cdot(|H|)^2$$

Equation 131

where

$$H=\cos(\gamma)^2 - \left(\frac{\sigma}{s_\beta}\right)\cdot\left(\frac{\sigma}{s_\alpha}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2 - i\cdot\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{s_\beta}+\frac{\sigma}{s_\alpha}\right)$$

Equation 132

so that

$$(|H|)^2=\left[\cos(\gamma)^2 - \left(\frac{\sigma}{s_\beta}\right)\cdot\left(\frac{\sigma}{s_\alpha}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 + \left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{s_\beta}+\frac{\sigma}{s_\alpha}\right)\right]^2$$

Equation 133

Meanwhile, the factor

$$\sin\left[\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]\cdot\sin\left[\left(\frac{\Delta k}{2}-i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]$$

may be expanded to

$$\sin\left[\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]\cdot\sin\left[\left(\frac{\Delta k}{2}-i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]=\sin\left(\frac{\Delta k\cdot\Lambda}{2}\right)^2\cdot\cosh\left(\frac{\Delta\alpha\cdot\Lambda}{4}\right)^2 \ldots + \cos\left(\frac{\Delta k\cdot\Lambda}{2}\right)^2\cdot\sinh\left(\frac{\Delta\alpha\cdot\Lambda}{4}\right)^2$$ Equation 134

and, for small induced birefringence and dichroism, approximated by

$$\sin\left[\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]\cdot\sin\left[\left(\frac{\Delta k}{2}-i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right]=\left(\frac{\Delta k\cdot\Lambda}{2}\right)^2+\left(\frac{\Delta\alpha\cdot\Lambda}{4}\right)^2$$ Equation 135

Remembering the relationship between the induced birefringence, Δn, and the induced dichroism, $\Delta\alpha$, the expression for the signal strength becomes

$$\frac{I_{probe\_transmitted\_analyser}}{I_{probe\_0}\cdot e^{-\alpha\cdot\Lambda}}=4\cdot\frac{(|H|)^2}{(\sigma\cdot\cos(\chi))^2}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha-s_\beta}\right)^2\cdot\left[\left(\frac{\Delta\alpha\cdot\Lambda}{4}\right)^2\cdot\frac{1}{1+x^2}\right]$$ Equation 136

or, as

$$\Delta n_0=\frac{1}{2}\cdot\Delta\alpha_0\cdot\frac{c}{\omega_0}$$ Equation 137

$$\frac{I_{probe\_transmitted\_analyser}}{I_{probe\_0}\cdot e^{-\alpha\cdot\Lambda}}=4\cdot\frac{(|H|)^2}{(\sigma\cdot\cos(\chi))^2}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha-s_\beta}\right)^2\cdot\left[\left(\frac{\Delta n_0\cdot\Lambda}{2}\right)^2\cdot\left(\frac{\omega_0}{c}\right)^2\cdot\frac{1}{1+x^2}\right]$$ Equation 138

Consolidating the geometric dependence of the signal strength (including the dependence inherent in the induced birefringence and the pump/probe beam interaction region) into the factor, $J(\gamma,\varphi)_{0\_approx}$, this becomes

$$\frac{I_{probe\_transmitted\_analyser}}{I_{probe\_0}\cdot e^{-\alpha\cdot\Lambda}}=\frac{4\cdot(|H|)^2}{(\sigma\cdot\cos(\chi))^2}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha-s_\beta}\right)^2\cdot\left[\left(\frac{n_\alpha-n_\beta}{n_o}\right)^2\cdot\left(\frac{\Lambda}{W}\right)^2\right]\cdot\left[\left(\frac{n_0\cdot\omega_0}{2\cdot c}\right)^2\cdot\frac{1}{1+x^2}\right]\cdot W^2$$

Equation 139

or

$$\frac{I_{probe\_transmitted\_analyser}}{I_{probe\_0}\cdot e^{-\alpha\cdot\Lambda}}=J(\gamma,\chi)_{0\_approx}\cdot\left[\left(\frac{n_0\cdot\omega_0}{2\cdot c}\right)^2\cdot\frac{1}{1+x^2}\right]\cdot W^2$$ Equation 140

where

$$J(\gamma,\chi)_{0\_approx}=\frac{4\cdot(|H|)^2}{(\sigma\cdot\cos(\chi))^2}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha-s_\beta}\right)^2\cdot\left(\frac{n_\alpha-n_\beta}{n_o}\right)^2\cdot\left(\frac{\Lambda}{W}\right)^2$$ Equation 141

$$J(\gamma,\chi)_{0\_approx}=\frac{4\cdot(|H|)^2}{(\sigma\cdot\cos(\chi))^2}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha-s_\beta}\right)^2\cdot\left(\frac{n_\alpha-n_\beta}{n_o}\right)^2\cdot\left(\frac{1}{\sin(\chi)^2}\right)$$ Equation 142

$$J(\gamma,\chi)_{0\_approx} = \frac{4\cdot(|H|)^2}{(\sigma\cdot\cos(\chi)\cdot\sin(\chi))^2}\cdot\left(\frac{s_\alpha\cdot s_\beta}{s_\alpha - s_\beta}\right)^2\cdot\left(\frac{n_\alpha - n_\beta}{n_o}\right)^2$$ Equation 143

Remembering that

$$\frac{n_\alpha - n_\beta}{n_o} = \sqrt{1+s_\alpha} - \sqrt{1+s_\beta} \sim 1 + \frac{s_\alpha}{2} - \left(1 + \frac{s_\beta}{2}\right) = \frac{s_\alpha - s_\beta}{2}$$ Equation 144

equation [143] becomes

$$J(\gamma,\chi)_{0\_approx} = \frac{(|H|)^2}{(\sigma\cdot\cos(\chi)\cdot\sin(\chi))^2}\cdot\left(s_\alpha\cdot s_\beta\right)^2$$ Equation 145

where

$$(|H|)^2 = \left[\cos(\gamma)^2 - \left(\frac{\sigma}{s_\beta}\right)\cdot\left(\frac{\sigma}{s_\alpha}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 + \left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{s_\beta} + \frac{\sigma}{s_\alpha}\right)\right]^2$$

Equation 146

Case 1: $\varphi = \chi \sim 0$

$$s_{\alpha\_0\_approx\_1} = \cos(\chi)\cdot\sigma$$ Equation 147

$$s_{\beta\_0\_approx\_1} = -\cos(\chi)\cdot\sigma$$ Equation 148

so that

$$\left[(|H|)^2\right]_{0\_approx\_1} = \left[\cos(\gamma)^2 - \left(\frac{\sigma}{-\cos(\chi)\cdot\sigma}\right)\cdot\left(\frac{\sigma}{\cos(\chi)\cdot\sigma}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 \ldots$$ Equation 149

$$+\left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{-\cos(\chi)\cdot\sigma} + \frac{\sigma}{\cos(\chi)\cdot\sigma}\right)\right]^2$$

$$\left[(|H|)^2\right]_{0\_approx\_1} = \left(\cos(\gamma)^2 + \sin(\gamma)^2\right)^2 = 1$$ Equation 150

and

$$J(\gamma,\chi)_{0\_approx\_1} = \frac{1}{(\sigma\cdot\cos(\chi)\cdot\sin(\chi))^2}\cdot\left(-\sigma^2\cdot\cos(\chi)^2\right)^2$$ Equation 151

$$J(\gamma,\chi)_{0\_approx\_1} = \sigma^2\cdot\cot(\chi)^2$$ Equation 152

Note that this expression is independent of the plane of polarisation of the probe beam defined by the angle, γ.

Case 2: $\varphi = \chi \sim \pi/2$, but with the proviso that $\cos^2(\chi) >> \sigma^2, \Delta$

$$s_{\alpha\_0\_approx\_2} = \Delta\cdot\sin(\chi)^2$$ Equation 153

$$s_{\beta\_0\_approx\_2} = -\sigma^2\cdot\sin(\chi)^2$$ Equation 154

so that

$$\left[(|H|)^2\right]_{0\_approx\_2} = \left[\cos(\gamma)^2 - \left(\frac{\sigma}{-\sigma^2\cdot\sin(\chi)^2}\right)\cdot\left(\frac{\sigma}{\Delta\cdot\sin(\chi)^2}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 \ldots + \left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{-\sigma^2\cdot\sin(\chi)^2} + \frac{\sigma}{\Delta\cdot\sin(\chi)^2}\right)\right]^2$$ Equation 155

$$\left[(|H|)^2\right]_{0\_approx\_2} = \left[\cos(\gamma)^2 + \left(\frac{1}{\Delta}\right)\cdot\frac{\cos(\chi)^2}{\sin(\chi)^4}\cdot\sin(\gamma)^2\right]^2 \ldots + \left[\frac{\cos(\chi)}{\sin(\chi)^2}\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{\Delta} - \frac{1}{\sigma}\right)\right]^2$$ Equation 156

and

$$J(\gamma,\chi)_{0\_approx\_2} = \frac{(|H|)^2}{(\sigma\cdot\cos(\chi)\cdot\sin(\chi))^2}\cdot\left[-\sigma^2\cdot\sin(\chi)^2\cdot\left(\Delta\cdot\sin(\chi)^2\right)\right]^2$$ Equation 157

$$J(\gamma,\chi)_{0\_approx\_2} = \left[\left[\cos(\gamma)^2 + \left(\frac{1}{\Delta}\right)\cdot\frac{\cos(\chi)^2}{\sin(\chi)^4}\cdot\sin(\gamma)^2\right]^2 \ldots + \left[\frac{\cos(\chi)}{\sin(\chi)^2}\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\frac{\sigma}{\Delta} - \frac{1}{\sigma}\right)\right]^2\right]\cdot\left(\sigma\cdot\Delta\cdot\frac{\sin(\chi)^3}{\cos(\chi)}\right)^2$$ Equation 158

Let us consider three simple cases of this expression.

If $\gamma = 0$, equation [158] becomes

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=0} = (\sigma)^2\cdot\Delta^2\cdot\frac{\sin(\chi)^6}{\cos(\chi)^2}$$ Equation 159

if $\gamma = \pi/2$, equation [158] becomes

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=\frac{\pi}{2}} = \left(\frac{1}{\Delta}\cdot\frac{\cos(\chi)^2}{\sin(\chi)^4}\right)^2\cdot\left(\sigma\cdot\Delta\cdot\frac{\sin(\chi)^3}{\cos(\chi)}\right)^2$$ Equation 160

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=\frac{\pi}{2}} = \left(\frac{\cos(\chi)}{\sin(\chi)}\right)^2\cdot\sigma^2$$ Equation 161

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=\frac{\pi}{2}} = \cot(\chi)^2\cdot\sigma^2$$ Equation 162

and if $\gamma = \pi/4$, equation [158] becomes

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=\frac{\pi}{4}} = \left[\left[\frac{1}{2} + \left(\frac{1}{\Delta}\right)\cdot\frac{\cos(\chi)^2}{\sin(\chi)^4}\cdot\frac{1}{2}\right]^2 \ldots + \left[\frac{\cos(\chi)}{\sin(\chi)^2}\cdot\left(\frac{1}{2}\right)\cdot\left(\frac{\sigma}{\Delta} - \frac{1}{\sigma}\right)\right]^2\right]\cdot\left(\sigma\cdot\Delta\cdot\frac{\sin(\chi)^3}{\cos(\chi)}\right)^2$$ Equation 163

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=\frac{\pi}{4}} = \frac{1}{4}\cdot\frac{\left[\left(\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)\cdot\sigma\right]^2 \ldots + \left[\cos(\chi)\cdot\sin(\chi)^2\cdot\left(\sigma^2 - \Delta\right)\right]^2}{(\sin(\chi)\cdot\cos(\chi))^2}$$ Equation 164

Consider also <u>the more general case</u>: all φ (= χ) such that $\cos^2(\varphi) >> \sigma^2, \Delta$.

The general s functions may be written in the form

$$s_{\alpha\_O\_approx} = a + \sqrt{b}$$ Equation 165

$$s_{\beta\_o\_approx} = a - \sqrt{b}$$ Equation 166

where

$$a = \frac{\Delta - \sigma^2}{2}\cdot\sin(\chi)^2$$ Equation 167

and

$$b = \sqrt{\sin(\chi)^4\cdot\left(\frac{\Delta + \sigma^2}{2}\right)^2 + \cos(\chi)^2\cdot\sigma^2}$$ Equation 168

so that

$$s_{\alpha\_0\_approx} + s_{\beta\_0\_approx} = 2\cdot a = \left(\Delta - \sigma^2\right)\cdot\sin(\chi)^2$$ Equation 169

$$s_{\alpha\_0\_approx}\cdot s_{\beta\_0\_approx} = a^2 - b = -\left[\left(\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)\cdot\sigma^2\right]$$ Equation 170

so that

$$\left[(|H|)^2\right]_{\_approx} = \left[\cos(\gamma)^2 - \left(\frac{\sigma^2}{a^2 - b}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 \ldots + \left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\sigma\cdot\left(\frac{2\cdot a}{a^2 - b}\right)\right]^2$$ Equation 171

and

$$J(\gamma,\chi)_{0\_approx} = \frac{\left[\cos(\gamma)^2 - \left(\frac{\sigma^2}{a^2-b}\right)\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 \ldots \\ + \left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\sigma\cdot\left(\frac{2\cdot a}{a^2-b}\right)\right]^2}{(\sigma\cdot\cos(\chi)\cdot\sin(\chi))^2}\cdot\left(a^2-b\right)^2$$ Equation 172

$$J(\gamma,\chi)_{0\_approx} = \frac{\left[\cos(\gamma)^2\cdot\left(\frac{a^2-b}{\sigma}\right) - \sigma\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 \ldots \\ + (\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot(2\cdot a))^2}{(\cos(\chi)\cdot\sin(\chi))^2}$$ Equation 173

$$J(\gamma,\chi)_{0\_approx} = \frac{\left[-\cos(\gamma)^2\cdot\left(\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)\cdot\sigma - \sigma\cdot\cos(\chi)^2\cdot\sin(\gamma)^2\right]^2 \ldots \\ + \left[\cos(\chi)\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left[\left(\Delta-\sigma^2\right)\cdot\sin(\chi)^2\right]\right]^2}{(\cos(\chi)\cdot\sin(\chi))^2}$$ Equation 174

$$J(\gamma,\chi)_{0\_approx} = \frac{\left[\left(\cos(\gamma)^2\cdot\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)\cdot\sigma\right]^2 \ldots \\ + \left[\cos(\chi)\cdot\sin(\chi)^2\cdot\cos(\gamma)\cdot\sin(\gamma)\cdot\left(\Delta-\sigma^2\right)\right]^2}{(\cos(\chi)\cdot\sin(\chi))^2}$$ Equation 175

If $\gamma = 0$, this becomes

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=0} = \frac{\left(\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)^2}{(\cos(\chi)\cdot\sin(\chi))^2}\cdot\sigma^2$$ Equation 176

which for $\chi \sim 0$ reduces to

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=0} = \cot(\chi)^2\cdot\sigma^2$$ Equation 177

and for $\chi \sim \pi/2$ (such that $\cos^2(\varphi) >> \sigma^2, \Delta$) does not reduce further and is given by

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=0} = \frac{\left(\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)^2}{(\cos(\chi)\cdot\sin(\chi))^2}\cdot\sigma^2$$ Equation 178

showing the general extension to the expression derived for case 2 and $\gamma \sim \pi/2$ in equation [159], i.e.

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=0} = \sigma^2\cdot\Delta^2\cdot\frac{\sin(\chi)^6}{\cos(\chi)^2}$$ Equation 179

If $\gamma = \pi/2$, equation [175] becomes

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=\frac{\pi}{2}} = \cot(\chi)^2 \cdot \sigma^2$$ Equation 180

for all values of χ, where $\cos^2(\chi) >> \sigma^2$, Δ.,

while for $\gamma = \pi/4$, equation [175] becomes

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=\frac{\pi}{2}} = \frac{1}{4} \cdot \frac{\left[\left(\Delta \cdot \sin(\chi)^4 + 2 \cdot \cos(\chi)^2\right) \cdot \sigma\right]^2 \ldots + \left[\cos(\chi) \cdot \sin(\chi)^2 \cdot \left(\Delta - \sigma^2\right)\right]^2}{(\cos(\chi) \cdot \sin(\chi))^2}$$ Equation 181

showing an extra factor of 2 in the first term in the numerator on the right hand side of the expression when compared with the general extension to the expression derived for case 2 and $\gamma = \pi/4$ in equation [164], due to the more accurate definition of the induced dichroism (equations [165] and [166]) in this case i.e.

$$\left(J(\gamma,\chi)_{0\_approx\_2}\right)_{\gamma=\frac{\pi}{2}} = \frac{1}{4} \cdot \frac{\left[\left(\Delta \cdot \sin(\chi)^4 + \cos(\chi)^2\right) \cdot \sigma\right]^2 \ldots + \left[\cos(\chi) \cdot \sin(\chi)^2 \cdot \left(\sigma^2 - \Delta\right)\right]^2}{(\sin(\chi) \cdot \cos(\chi))^2}$$ Equation 182

Note the derivation above is correct only for $\cos^2(\chi) >> \sigma^2$, Δ.

<u>We now consider the extreme of case 2</u>: $\varphi = \chi \sim \pi/2$, $\cos^2(\varphi) << \sigma^2$, Δ. The treatment is more complex as the electric field components may not be expressed in forms clearly identifiable as lying in the plane of polarisation of the probe beam.

Remembering to swap the X and Y components to match the polarisation spectroscopy geometry, the electric field vectors may be written

$$E_{\alpha\_0\_approx\_2} = \Delta \cdot \sin(\varphi)^2 \cdot \begin{bmatrix} i \cdot \left(\frac{\sigma}{\Delta}\right) \cdot \frac{\cos(\varphi)^2}{\sin(\varphi)^2} \\ \cos(\varphi)^2 \\ -\sin(\varphi) \cdot \cos(\varphi) \end{bmatrix}$$ Equation 183

$$E_{\beta\_0\_approx\_2} = -\sigma \cdot \sin(\varphi)^2 \cdot \begin{bmatrix} i \cdot \left(\frac{\cos(\varphi)^2 + \sigma^2 + \Delta}{\sin(\varphi)^2}\right) \\ \left(\cos(\varphi)^2 + \sigma^2 + \Delta\right) \cdot \sigma \\ -\sin(\varphi) \cdot \cos(\varphi) \cdot \sigma \end{bmatrix}$$ Equation 184

and approximated as, since $\cos^2(\chi) << \sigma^2$, Δ.

$$E_{\alpha\_0\_approx\_2} = \Delta \cdot \sin(\varphi)^2 \cdot \cos(\varphi) \cdot \begin{bmatrix} i \cdot \left(\frac{\sigma}{\Delta}\right) \cdot \frac{\cos(\varphi)}{\sin(\varphi)^2} \\ \cos(\varphi) \\ -\sin(\varphi) \end{bmatrix}$$ Equation 185

and

$$E_{\beta\_0\_approx\_2} = -\sigma \cdot \sin(\varphi)^2 \cdot \begin{bmatrix} i \cdot \left(\frac{\sigma^2 + \Delta}{\sin(\varphi)^2}\right) \\ \left(\sigma^2 + \Delta\right) \cdot \sigma \\ -\sin(\varphi) \cdot \cos(\varphi) \cdot \sigma \end{bmatrix}$$ Equation 186

or

$$E_{\beta\_0\_approx\_2} = -\sigma \cdot \sin(\varphi)^2 \cdot i \cdot \left(\sigma^2 + \Delta\right) \cdot \begin{bmatrix} \left(\frac{1}{\sin(\varphi)^2}\right) \\ -i \cdot \sigma \\ i \cdot \sin(\varphi) \cdot \cos(\varphi) \cdot \frac{\sigma}{\sigma^2 + \Delta} \end{bmatrix}$$ Equation 187

In the second case, the α and β electric field components approach (as $\chi \sim \pi/2$)

$$E_{\alpha\_0\_approx\_2} = \Delta \cdot \sin(\varphi)^2 \cdot \cos(\varphi) \cdot \begin{bmatrix} 0 \\ 0 \\ -1 \end{bmatrix}$$ Equation 188

and

$$E_{\beta\_0\_approx\_2} = -\sigma \cdot \sin(\varphi)^2 \cdot i \cdot \left(\sigma^2 + \Delta\right) \cdot \begin{bmatrix} 1 \\ -i \cdot \sigma \\ 0 \end{bmatrix}$$ Equation 189

The α component polarisation mode lies primarily aligned to the Z axis, while the β component lies primarily along the X axis (the Y axis in the original diagonalised geometry). We may approximate a first solution to equation [110], in the limit of $\cos^2(\chi) << \sigma^2, \Delta$ by normalising the vectors to a maximum value of unity along these axes when $\chi = \pi/2$ and setting the complex electric field components to zero to describe the transmitted probe beam electric field as the propagation of purely linearly polarised probe beam components under the action of the induced birefringence due to the optical activity.

Working from equations [164] and [163], we find the approximate linearly polarised solutions to be

$$E_{\alpha\_0\_approx\_2\_linear} = \begin{bmatrix} 0 \\ \dfrac{\cos(\chi)}{\sin(\chi)} \\ -1 \end{bmatrix} \text{ or, renormalised to } \begin{bmatrix} 0 \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix} \qquad \text{Equation 190}$$

and

$$E_{\beta\_0\_approx\_2\_linear} = \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix} \qquad \text{Equation 191}$$

As a result, we assume the YZ probe components of the probe beam are subject to the α refractive index solution, and the X component of the probe beam is subject to the β refractive index solution and rewrite equation [110] as

$$\begin{bmatrix} \cos(\gamma) \\ \sin(\gamma)\cdot\cos(\chi) \\ -\sin(\gamma)\cdot\sin(\chi) \end{bmatrix} = \begin{bmatrix} 0 \\ \sin(\gamma)\cdot\cos(\chi) \\ -\sin(\gamma)\cdot\sin(\chi) \end{bmatrix} + \begin{bmatrix} \cos(\gamma) \\ 0 \\ 0 \end{bmatrix} \qquad \text{Equation 192}$$

or

$$\begin{bmatrix} \cos(\gamma) \\ \sin(\gamma)\cdot\cos(\chi) \\ -\sin(\gamma)\cdot\sin(\chi) \end{bmatrix} = \sin(\gamma)\cdot\begin{bmatrix} 0 \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix} + \cos(\gamma)\cdot\begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix} \qquad \text{Equation 193}$$

After passage through a distance, Λ, through the dichroic and birefringent region, the probe beam is

$$\frac{E_{probe\_transmitted\_\ \Lambda ... + linear}}{E_{probe\_0}} = \sin(\gamma)\cdot\begin{bmatrix} 0 \\ \cos(\chi) \\ -\sin(\chi) \end{bmatrix}\cdot e^{-\frac{\alpha_\alpha}{2}\cdot\Lambda + i\cdot k_\alpha\cdot\Lambda} + \cos(\gamma)\cdot\begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix}\cdot e^{-\frac{\alpha_\beta}{2}\cdot\Lambda + i\cdot k_\beta\cdot\Lambda}$$

Equation 194

or

$$\frac{E_{probe\_transmitted\_\ \Lambda ... + linear}}{E_{probe\_0}} = \begin{bmatrix} \cos(\gamma)\cdot e^{-\frac{\alpha_\beta}{2}\cdot\Lambda + i\cdot k_\beta\cdot\Lambda} \\ \cos(\chi)\cdot\sin(\gamma)\cdot e^{-\frac{\alpha_\alpha}{2}\cdot\Lambda + i\cdot k_\alpha\cdot\Lambda} \\ -\sin(\chi)\cdot\sin(\gamma)\cdot e^{-\frac{\alpha_\alpha}{2}\cdot\Lambda + i\cdot k_\alpha\cdot\Lambda} \end{bmatrix} \qquad \text{Equation 195}$$

Letting

$$\alpha_{\alpha} = \alpha + \frac{\Delta\alpha}{2} \qquad k_{\alpha} = k + \frac{\Delta k}{2}$$ Equation 196

$$\alpha_{\beta} = \alpha - \frac{\Delta\alpha}{2} \qquad k_{\beta} = k - \frac{\Delta k}{2}$$ Equation 197

the transmitted probe beam electric field is

$$\frac{E_{probe\_transmitted\_\Lambda ...+linear}}{E_{probe\_0}} = \begin{bmatrix} \cos(\gamma)\cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda} \\ \sin(\gamma)\cdot\cos(\chi)\cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \\ -\sin(\gamma)\cdot\sin(\chi)\cdot e^{\frac{-\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \end{bmatrix} \cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$ Equation 198

The fraction of the probe beam transmitted through the crossed analyser is then given by

$$\frac{E_{probe\_transmitted ...+linear}}{E_{probe\_0}} = \begin{bmatrix} \cos(\gamma)\cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda} \\ \sin(\gamma)\cdot\cos(\chi)\cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \\ -\sin(\gamma)\cdot\sin(\chi)\cdot e^{\frac{-\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} \end{bmatrix} \cdot \begin{bmatrix} -\sin(\gamma) \\ \cos(\gamma)\cdot\cos(\chi) \\ -\cos(\gamma)\cdot\sin(\chi) \end{bmatrix} \cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$

Equation 199

$$\frac{E_{probe\_transmitted ...+linear}}{E_{probe\_0}} = \begin{bmatrix} \left(\cos(\chi)^2 + \sin(\chi)^2\right)\cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} ... \\ + -1\cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda} \end{bmatrix} \cdot \sin(\gamma)\cdot\cos(\gamma)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$

Equation 200

$$\frac{E_{probe\_transmitted ...+linear}}{E_{probe\_0}} = \left(e^{-\frac{\Delta\alpha}{4}\cdot\Lambda + i\cdot\frac{\Delta k}{2}\cdot\Lambda} - e^{\frac{\Delta\alpha}{4}\cdot\Lambda - i\cdot\frac{\Delta k}{2}\cdot\Lambda}\right) \cdot \sin(\gamma)\cdot\cos(\gamma)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$ Equation 201

$$\frac{E_{probe\_transmitted ...+linear}}{E_{probe\_0}} = \left[e^{i\cdot\left(\frac{\Delta k}{2} + i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda} - e^{-i\cdot\left(\frac{\Delta k}{2} + i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda} \right] \cdot \sin(\gamma)\cdot\cos(\gamma)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$

Equation 202

or

$$\frac{E_{probe\_transmitted ...+linear}}{E_{probe\_0}} = 2i\cdot\sin\left[\left(\frac{\Delta k}{2} + i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda\right] \cdot \sin(\gamma)\cdot\cos(\gamma)\cdot e^{-\frac{\alpha}{2}\cdot\Lambda} \cdot e^{i\cdot k\cdot\Lambda}$$ Equation 203

This corresponds to the transmitted probe beam intensity

$$\frac{I_{probe\_transmitted\ ...\ +linear}}{I_{probe\_0}} = 4 \cdot \left[\sin\left[\left(\frac{\Delta k}{2} + i \cdot \frac{\Delta\alpha}{4} \right) \cdot \Lambda \right] \cdot \sin\left[\left(\frac{\Delta k}{2} - i \cdot \frac{\Delta\alpha}{4} \right) \cdot \Lambda \right] \right] \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot e^{-\alpha \cdot \Lambda}$$

Equation 204

or

$$\frac{I_{probe\_transmitted\ ...\ +linear}}{I_{probe\_0}} = 4 \cdot \left[\sin\left(\frac{\Delta k}{2} \cdot \Lambda \right)^2 \cdot \cosh\left(\frac{\Delta\alpha}{4} \cdot \Lambda \right)^2 ... + \cos\left(\frac{\Delta k}{2} \cdot \Lambda \right)^2 \cdot \sinh\left(\frac{\Delta\alpha}{4} \cdot \Lambda \right)^2 \right] \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot e^{-\alpha \cdot \Lambda}$$

Equation 205

For small induced dichroism and birefringence, this may be further approximated to

$$\frac{I_{probe\_transmitted\ ...\ +linear}}{I_{probe\_0}} = 4 \cdot \left[\left(\frac{\Delta k}{2} \cdot \Lambda \right)^2 + \left(\frac{\Delta\alpha}{4} \cdot \Lambda \right)^2 \right] \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot e^{-\alpha \cdot \Lambda}$$

Equation 206

Including the Λ and Δn dependence as for the case of $\cos^2(\chi) >> \sigma^2, \Delta$, the geometric dependence of the polarisation spectroscopy signal strength for a circularly polarised pump beam for $\varphi \sim \pi/2$ is then proportional to the factor (as defined in equation [140] above)

$$J(\gamma,\chi)_{0\_approx\_2\ ...\ +linear} = 4 \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot \left(\frac{n_\alpha - n_\beta}{n_o} \right)^2 \cdot \left(\frac{\Lambda}{W} \right)^2$$

Equation 207

$$J(\gamma,\chi)_{0\_approx\_2\ ...\ +linear} = 4 \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot \left[\left(\frac{\Delta + \sigma^2}{2} \right) \cdot \sin(\chi)^2 \right]^2 \cdot \left(\frac{1}{\sin(\chi)} \right)^2$$

Equation 208

$$J(\gamma,\chi)_{0\_approx\_2\ ...\ +linear} = 4 \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot \sin(\chi)^2 \cdot \left(\frac{\Delta + \sigma^2}{2} \right)^2$$

Equation 209

For $\gamma = 0$, this reduces to

$$\left(J(\gamma,\chi)_{0\_approx\_2\ ...\ +linear} \right)_{\gamma=0} = 0$$

Equation 210

for $\gamma = \pi/2$, to

$$\left(J(\gamma,\chi)_{0\_approx\_2\ ...\ +linear} \right)_{\gamma=\frac{\pi}{2}} = 0$$

Equation 211

and for $\gamma = \pi/4$, to

$$\left(J(\gamma,\chi)_{\substack{0\_approx\_2\ ...\\ +\,linear}}\right)_{\gamma=\frac{\pi}{4}} = \sin(\chi)^2 \cdot \left(\frac{\Delta + \sigma^2}{2}\right)^2$$

Equation 212

In Summary:

The polarisation spectroscopy signal strength dependence on the intersection angle of pump and probe beams, χ, is given by

For $\gamma = 0$

For $\cos^2(\chi) >> \sigma^2, \Delta$

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=0} = \frac{\left(\Delta\cdot\sin(\chi)^4 + \cos(\chi)^2\right)^2}{(\cos(\chi)\cdot\sin(\chi))^2}\cdot\sigma^2 = \left(\Delta\cdot\sin(\chi)^2\cdot\tan(\chi) + \cot(\chi)\right)^2\cdot\sigma^2$$

Equation 213

which tends to infinity as $\cos(\chi)$ approaches zero.

For $\cos^2(\chi) << \sigma^2, \Delta$

$$\left(J(\gamma,\chi)_{\substack{0\_approx\_2\ ...\\ +\,linear}}\right)_{\gamma=0} = 0$$

Equation 214

For $\gamma = \pi/2$, the signal strength dependence is given by

For $\cos^2(\chi) >> \sigma^2, \Delta$

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=0} = \cot(\chi)^2\cdot\sigma^2$$

Equation 215

which tends to zero as $\cos(\chi)$ approaches zero.

For $\cos^2(\chi) << \sigma^2, \Delta$

$$\left(J(\gamma,\chi)_{\substack{0\_approx\_2\ ...\\ +\,linear}}\right)_{\gamma=\frac{\pi}{2}} = 0$$

Equation 216

For $\gamma = \pi/4$, the signal strength dependence is given by

For $\cos^2(\chi) >> \sigma^2, \Delta$

$$\left(J(\gamma,\chi)_{0\_approx}\right)_{\gamma=\frac{\pi}{2}} = \frac{1}{4}\cdot\frac{\left[\left(\Delta\cdot\sin(\chi)^4 + 2\cdot\cos(\chi)^2\right)\cdot\sigma\right]^2 \ ... + \left[\cos(\chi)\cdot\sin(\chi)^2\cdot\left(\Delta - \sigma^2\right)\right]^2}{(\cos(\chi)\cdot\sin(\chi))^2} = \frac{1}{4}\cdot\left[\left[\left(\Delta\cdot\sin(\chi)^2\cdot\tan(\chi) + 2\cdot\cot(\chi)\right)\cdot\sigma\right]^2 \ ... + \left[\sin(\chi)\cdot\left(\Delta - \sigma^2\right)\right]^2\right]$$

Equation 217

which tends to infinity as $\cos(\chi)$ approaches zero due to the $\tan(\chi)$ function in the first term..

For $\cos^2(\chi) << \sigma^2, \Delta$

$$\left(J(\gamma,\chi)_{\substack{0\_approx\_2\ \ldots \\ +linear}}\right)_{\gamma=\frac{\pi}{4}} = \sin(\chi)^2 \cdot \left(\frac{\Delta + \sigma^2}{2}\right)^2$$ Equation 218

These expression predicts zero signal strength for orthogonal pump/probe beam intersection is obtained for probe beam polarisation angles of $\gamma = 0, \pi/2$.

The general signal strength dependence is given by

For $\cos^2(\chi) >> \sigma^2, \Delta$

$$J(\gamma,\chi)_{0\_approx} = \frac{\left[\left(\cos(\gamma)^2 \cdot \Delta \cdot \sin(\chi)^4 + \cos(\chi)^2\right) \cdot \sigma\right]^2 \ldots + \left[\cos(\chi) \cdot \sin(\chi)^2 \cdot \cos(\gamma) \cdot \sin(\gamma) \cdot \left(\Delta - \sigma^2\right)\right]^2}{\left(\cos(\chi) \cdot \sin(\chi)\right)^2}$$ Equation 219

or

$$J(\gamma,\chi)_{0\_approx} = \left[\left(\cos(\gamma)^2 \cdot \Delta \cdot \sin(\chi)^2 \cdot \tan(\chi) + \cot(\chi)\right) \cdot \sigma\right]^2 \ldots + \left[\sin(\chi) \cdot \cos(\gamma) \cdot \sin(\gamma) \cdot \left(\Delta - \sigma^2\right)\right]^2$$ Equation 220

For $\cos^2(\chi) << \sigma^2, \Delta$

$$J(\gamma,\chi)_{\substack{0\_approx\_2\ \ldots \\ +linear}} = 4 \cdot \sin(\gamma)^2 \cdot \cos(\gamma)^2 \cdot \sin(\chi)^2 \cdot \left(\frac{\Delta + \sigma^2}{2}\right)^2$$ Equation 221

or

$$J(\gamma,\chi)_{\substack{0\_approx\_2\ \ldots \\ +linear}} = \sin(2 \cdot \gamma)^2 \cdot \sin(\chi)^2 \cdot \left(\frac{\Delta + \sigma^2}{2}\right)^2$$ Equation 222

Appendix VII: Calculation of the Additional Clebsch-Gordon Coefficient Sums

The algebraic expressions representing the Clebsch-Gordon coefficients used in this thesis are those quoted in Zare *"Angular Momentum"* (p 57, Table 2.4 C: $j_2 = 1$)[A1]. The squares of the Clebsch-Gordon coefficients represent probabilities, while the Clebsch-Gordon coefficients themselves represent probability amplitudes.

Note that the selection rules for the Clebsch-Gordon coefficients require that the combined state magnetic quantum number, m, represents the algebraic sum of the two component magnetic quantum numbers:

$m = m_1 + m_2$ Equation 1

The combined rotational quantum number, j, is the vector sum of the two component rotational quantum numbers:

$|j_1 + j_2| \geq j \geq |j_1 - j_2|$ Equation 2

This requires that the Clebsch-Gordon coefficients for P ($\Delta j = -1$) transitions are zero for $j_1 = 0$ and ½ and Q ($\Delta j = 0$) transitions are zero for $j_1 = 0$ for absorption or emission of a photon ($j_2 = 1$).

For convenience, we rewrite these equations in terms of the magnetic quantum number of the lower state, m_1, of the transition. The restricted selection rules quoted above are stated directly in the following expressions. To avoid unnecessary subscripts in the following derivation, we represent the initial quantum state, (j_1,m_1), of the transition as (J,M). This is not to be confused with the quantum state of the combined system, (j,m).

Note that the right circularly polarised Clebsch-Gordon coefficients are equivalent, on replacement of M by -M, to the left circularly polarised Clebsch-Gordon coefficients.

Left circularly polarised transitions (M = m - 1, m = M + 1)

R transition $\langle J\ M, 1\ 1 \mid J+1\ M+1\rangle = \sqrt{\dfrac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$ Equation 3

Q transition $\langle J\ M, 1\ 1 \mid J\ M+1\rangle = \mathrm{if}\left[J=0, 0, -\sqrt{\dfrac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}}\right]$ Equation 4

P transition $\langle J\ M, 1\ 1 \mid J-1\ M+1\rangle = \mathrm{if}\left[J<1, 0, \sqrt{\dfrac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}}\right]$ Equation 5

Linearly polarised transitions (M = m)

R transition $$\langle J\ \ M,\ 1\ \ 0 \mid J{+}1\ M\rangle = \sqrt{\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}}$$ Equation 6

Q transition $$\langle J\ \ M,\ 1\ \ 0 \mid J\ M\rangle = \mathrm{if}\left[J=0,0,\frac{M}{\sqrt{J\cdot(J+1)}}\right]$$ Equation 7

P transition $$\langle J\ \ M,\ 1\ \ 0 \mid J{-}1\ M\rangle = \mathrm{if}\left[J<1,0,-\sqrt{\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}}\right]$$ Equation 8

Right circularly polarised transitions (M = m + 1, m = M - 1)

R transition $$\langle J\ \ M,\ 1\ \ {-1} \mid J{+}1\ M-1\rangle = \sqrt{\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$ Equation 9

Q transition $$\langle J\ \ M,\ 1\ \ {-1} \mid J\ M-1\rangle = \mathrm{if}\left[j_1=0,0,\sqrt{\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}}\right]$$ Equation 10

P transition $$\langle J\ \ M,\ 1\ \ {-1} \mid J{-}1\ \ M-1\rangle = \mathrm{if}\left[j_1<1,0,\sqrt{\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}\right]$$ Equation 11

In addition, we include the Clebsch-Gordon coefficients for the case of the orthogonal probe beam component discussed in Appendix I ($M = m \pm 1$, $m = M \mp 1$)

R transition

$$(1/2)\left[\langle j_1\ \ m_1,\ 1\ \ {-1} \mid j_1{+}1\ m_1-1\rangle + \langle j_1\ \ m_1,\ 1\ \ 1 \mid j_1{+}1\ m_1{+}1\rangle\right] = \sqrt{\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}}$$

Equation 12

Q transition

$$(1/2)\left[\langle j_1\ \ m_1,\ 1\ \ {-1} \mid j_1\ m_1-1\rangle + \langle j_1\ \ m_1,\ 1\ \ 1 \mid j_1\ m_1{+}1\rangle\right] = \sqrt{\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}}$$ Equation 13

P transition

$$(1/2)\left[\langle j_1\ \ m_1,\ 1\ \ {-1} \mid j_1{-}1\ \ m_1-1\rangle + \langle j_1\ \ m_1,\ 1\ \ 1 \mid j_1{-}1\ \ m_1{+}1\rangle\right] = \sqrt{\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]}$$

Equation 14

The following sections calculate firstly the additional $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions defined in Chapter IV. The summations are required to calculate the additional induced dichroisms not described in Teets, Kowalski, Hill, Carlson and Hansch's collinear pump/probe beam theory. All calculations for this Appendix were determined using Mathcad Plus 5.0.

Calculation of the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions

The derivations in this appendix mirror those of Appendix I. The additional $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions calculated represent

- the linear dichroism induced by a circularly polarised pump beam, and
- the circular dichroism induced by a linearly polarised pump beam.

For simplicity, the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions of Appendix I are recalculated with linear pump beam polarisation states in the M-sums replaced by circularly polarised pump beam descriptions (to calculate the linear dichroism induced by a circularly polarised pump beam) and circular pump beam polarisation states replaced by linearly polarised pump beam descriptions (to calculate the circular dichroism induced by a linearly polarised pump beam). We make the same assumptions as for the $Z_{J,J',J''}$ functions of Appendix I.

To avoid excessive complication, the conditions represented by the restricted selection rules defined in equation [2] are discussed after the derivation of the more general expressions below.

Circular Dichroism Induced by a Linearly Polarised Pump Beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta\_add_{J,J+1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}\cdot\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ldots + -\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$\zeta\_add_{J,J+1,J+1} = 0$ Equation 15

$Z\_add_{J,J+1,J+1} = 0$ Equation 16

R (probe), Q (pump)

$$\zeta\_add_{J,J,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{M^2}{J\cdot(J+1)}\right]\cdot\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ldots + -\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{M^2}{J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$\zeta\_add_{J,J,J+1} = 0$ Equation 17

$Z\_add_{J,J,J+1} = 0$ Equation 18

R (probe), P (pump)

$$\zeta\_add_{J,J-1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\ldots + -\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}}$$

$\zeta\_add_{J,J-1,J+1} = 0$ Equation 19

$Z\_add_{J,J-1,J+1} = 0$ Equation 20

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta\_add_{J,J+1,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}\cdot\left[\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\ldots+-(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]}{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}}$$

$\zeta\_add_{J,J+1,J}=0$ Equation 21

$Z\_add_{J,J+1,J}=0$ Equation 22

Q (probe), Q (pump)

$$\zeta\_add_{J,J,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{M^2}{J\cdot(J+1)}\right]\cdot\left[\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\ldots+-(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]}{\sum_{M=-J}^{J}\frac{M^2}{J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}}$$

$\zeta\_add_{J,J,J}=0$ Equation 23

$Z\_add_{J,J,J}=0$ Equation 24

Q (probe), P (pump)

$$\zeta\_add_{J,J-1,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}\ldots+-(-1)^2\cdot\frac{(J-M)\cdot(J+M+1)}{2\cdot J\cdot(J+1)}\right]}{\sum_{M=-J}^{J}\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J-M+1)}{2\cdot J\cdot(J+1)}}$$

$\zeta\_add_{J,J-1,J}=0$ Equation 25

$Z\_add_{J,J-1,J}=0$ Equation 26

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta\_add_{J,J+1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}\right]\cdot\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\ldots + -\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M+1)}{(2\cdot J+1)\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$\zeta\_add_{J,J+1} = 0$ Equation 27

$Z\_add_{J,J+1,J-1} = 0$ Equation 28

P (probe), Q (pump)

$$\zeta\_add_{J,J,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{M^2}{J\cdot(J+1)}\right]\cdot\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\ldots + -\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{M^2}{J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$\zeta\_add_{J,J,J-1} = 0$ Equation 29

$Z\_add_{J,J,J-1} = 0$ Equation 30

P (probe), P (pump)

$$\zeta\_add_{J,J-1,J-1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\ldots + -\frac{(J-M)\cdot(J-M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M)\cdot(J+M)}{J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}}$$

$\zeta\_add_{J,J-1,J-1} = 0$ Equation 31

$Z\_add_{J,J-1,J-1} = 0$ Equation 32

The circular dichroism induced by a linearly polarised pump beam in the linear pumping regime is zero.

Linear Dichroism Induced by a Right Circularly Polarised Pump Beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta\_add_{J,J+1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J+1,J+1} = \frac{-3}{20}\cdot\frac{(2\cdot J-1)}{(2\cdot J+3)}\cdot\frac{J}{(J+1)} = \frac{-\zeta_{J,J+1,J+1}}{2}$$ Equation 33

$$Z\_add_{J,J+1,J+1} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J+1}}{C_{J,J+1}\cdot C_{J,J+1}}\cdot\zeta\_add_{J,J+1,J+1}$$

$$Z\_add_{J,J+1,J+1} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{20}\cdot(2\cdot J-1)\cdot\frac{J}{((2\cdot J+3)\cdot(J+1))}\right]$$

$$Z\_add_{J,J+1,J+1} = \frac{-1}{60}\cdot(2\cdot J+3)\cdot(2\cdot J-1)\cdot\frac{J}{\left[(2\cdot J+1)^2\cdot(J+1)\right]} = \frac{-Z_{J,J+1,J+1}}{2}$$ Equation 34

R (probe), Q (pump)

$$\zeta\_add_{J,J,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}\right]\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J,J+1} = \frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-\zeta_{J,J,J+1}}{2}$$ Equation 35

$$Z\_add_{J,J,J+1} = \frac{\sigma_{J,J}\cdot\sigma_{J,J+1}}{C_{J,J}\cdot C_{J,J+1}}\cdot\zeta\_add_{J,J,J+1} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)}\right]$$

$$Z\_add_{J,J,J+1} = \frac{1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-Z_{J,J,J+1}}{2}$$ Equation 36

R (probe), P (pump)

$$\zeta\_add_{J,J-1,J+1} = (2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J-1,J+1} = \frac{-3}{20} = \frac{-\zeta_{J,J-1,J+1}}{2}$$

Equation 37

$$Z\_add_{J,J-1,J+1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J+1}}{C_{J,J-1}\cdot C_{J,J+1}}\cdot\zeta\_add_{J,J-1,J+1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{20}\right)$$

$$Z\_add_{J,J-1,J+1} = \frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3) = \frac{-Z_{J,J-1,J+1}}{2}$$

Equation 38

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta\_add_{J,J+1,J} = (2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta\_add_{J,J+1,J} = \frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-\zeta_{J,J+1,J}}{2}$$

Equation 39

$$Z\_add_{J,J+1,J} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J}}{C_{J,J+1}\cdot C_{J,J}}\cdot\zeta\_add_{J,J+1,J} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)}\right]$$

$$Z\_add_{J,J+1,J} = \frac{1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-Z_{J,J+1,J}}{2}$$

Equation 40

Q probe , Q pump

$$\zeta\_add_{J,J,J} = (2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}\right]\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta\_add_{J,J,J} = \frac{-3}{20}\cdot\frac{\left(4\cdot J^2+4\,J-3\right)}{(J\cdot(J+1))} = \frac{-\zeta_{J,J,J}}{2}$$ Equation 41

$$Z\_add_{J,J,J} = \frac{\sigma_{J,J}\cdot\sigma_{J,J}}{C_{J,J}\cdot C_{J,J}}\cdot\zeta\_add_{J,J,J} = \left(\frac{1}{3}\right)\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{-3}{20}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))}\right]$$

$$Z\_add_{J,J,J} = \frac{-1}{60}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))} = \frac{-Z_{J,J,J}}{2}$$ Equation 42

Q (probe), P (pump)

$$\zeta\_add_{J,J-1,J} = (2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta\_add_{J,J-1,J} = \frac{3}{20}\cdot\frac{(2\cdot J+3)}{J} = \frac{-\zeta_{J,J-1,J}}{2}$$ Equation 43

$$Z\_add_{J,J-1,J} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J}}{C_{J,J-1}\cdot C_{J,J}}\cdot\zeta\_add_{J,J-1,J} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}\right]$$

$$Z\_add_{J,J-1,J} = \frac{1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J} = \frac{-Z_{J,J-1,J}}{2}$$ Equation 44

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta\_add_{J,J+1,J-1}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{(J^2+M^2-J)}{(J\cdot(2\cdot J+1))}\right]\right]}{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J-M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J+1,J-1}=\frac{-3}{20}=\frac{-\zeta_{J,J+1,J-1}}{2}$$ Equation 45

$$Z\_add_{J,J+1,J-1}=\frac{\sigma_{J,J+1}\cdot\sigma_{J,J-1}}{C_{J,J+1}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J+1,J-1}$$

$$Z\_add_{J,J+1,J-1}=\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{20}\right)$$

$$Z\_add_{J,J+1,J-1}=\frac{-1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)^2}\cdot(2\cdot J-1)=\frac{-Z_{J,J+1,J-1}}{2}$$ Equation 46

P (probe), Q (pump)

$$\zeta\_add_{J,J,J-1}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}\right]\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{(J^2+M^2-J)}{(J\cdot(2\cdot J+1))}\right]\right]}{\sum_{M=-J}^{J}\frac{(J-M+1)\cdot(J+M)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J,J-1}=\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}=\frac{-\zeta_{J,J,J-1}}{2}$$ Equation 47

$$Z\_add_{J,J,J-1}=\frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J,J-1}=\left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}\right]$$

$$Z\_add_{J,J,J-1}=\frac{1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J}=\frac{-Z_{J,J,J-1}}{2}$$ Equation 48

P (probe), P (pump)

$$\zeta\_add_{J,J-1,J-1}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\right]\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]\right]}{\sum_{M=-J}^{J}\frac{(J+M)\cdot(J+M-1)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J-1,J-1}=\frac{-3}{20}\cdot\frac{\left(\left(2\cdot J^2+5\cdot J+3\right)\right)}{((2\cdot J-1)\cdot J)}$$

$$\zeta\_add_{J,J-1,J-1}=\frac{-3}{20}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)}=\frac{-\zeta_{J,J-1,J-1}}{2}$$ Equation 49

$$Z\_add_{J,J-1,J-1}=\frac{\sigma_{J,J-1}\cdot\sigma_{J,J-1}}{C_{J,J-1}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J-1,J-1}$$

$$Z\_add_{J,J-1,J-1}=\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{20}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)}\right]$$

$$Z\_add_{J,J-1,J-1}=\frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)\cdot\frac{(J+1)}{J}=\frac{-Z_{J,J-1,J-1}}{2}$$ Equation 50

Linear Dichroism Induced by a Left Circularly Polarised Pump Beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta\_add_{J,J+1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J+1,J+1} = \frac{-3}{20}\cdot(2\cdot J-1)\cdot\frac{J}{((2\cdot J+3)\cdot(J+1))} = \frac{-\zeta_{J,J+1,J+1}}{2}$$ Equation 51

$$Z\_add_{J,J+1,J+1} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J+1}}{C_{J,J+1}\cdot C_{J,J+1}}\cdot\zeta\_add_{J,J+1,J+1}$$

$$Z\_add_{J,J+1,J+1} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{20}\cdot(2\cdot J-1)\cdot\frac{J}{((2\cdot J+3)\cdot(J+1))}\right]$$

$$Z\_add_{J,J+1,J+1} = \frac{-1}{60}\cdot(2\cdot J+3)\cdot(2\cdot J-1)\cdot\frac{J}{\left[(2\cdot J+1)^2\cdot(J+1)\right]} = \frac{-Z_{J,J+1,J+1}}{2}$$ Equation 52

R (probe), Q (pump)

$$\zeta\_add_{J,J,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}\right]\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J,J+1} = \frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-\zeta_{J,J,J+1}}{2}$$ Equation 53

$$Z\_add_{J,J,J+1} = \frac{\sigma_{J,J}\cdot\sigma_{J,J+1}}{C_{J,J}\cdot C_{J,J+1}}\cdot\zeta\_add_{J,J,J+1} = \left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)}\right]$$

$$Z\_add_{J,J,J+1} = \frac{1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-Z_{J,J,J+1}}{2}$$ Equation 54

R (probe), P (pump)

$$\zeta\_add_{J,J-1,J+1} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\frac{\left(J^2+3\cdot J+M^2+2\right)}{((2\cdot J+1)\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{(J+1)^2-M^2}{(J+1)\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J-1,J+1} = \frac{-3}{20} = \frac{-\zeta_{J,J-1,J+1}}{2} \qquad \text{Equation 55}$$

$$Z\_add_{J,J-1,J+1} = \frac{\sigma_{J,J-1}\cdot\sigma_{J,J+1}}{C_{J,J-1}\cdot C_{J,J+1}}\cdot\zeta\_add_{J,J-1,J+1} = \left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{20}\right)$$

$$Z\_add_{J,J-1,J+1} = \frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3) = \frac{-Z_{J,J-1,J+1}}{2} \qquad \text{Equation 56}$$

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta\_add_{J,J+1,J} = (2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\right]\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta\_add_{J,J+1,J} = \frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-\zeta_{J,J+1,J}}{2} \qquad \text{Equation 57}$$

$$Z\_add_{J,J+1,J} = \frac{\sigma_{J,J+1}\cdot\sigma_{J,J}}{C_{J,J+1}\cdot C_{J,J}}\cdot\zeta\_add_{J,J+1,J} = \frac{1}{3}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)}\right]$$

$$Z\_add_{J,J+1,J} = \frac{1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-Z_{J,J+1,J}}{2} \qquad \text{Equation 58}$$

Q probe , Q pump

$$\zeta\_add_{J,J,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}\right]\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta\_add_{J,J,J}=\frac{-3}{20}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))}=\frac{-\zeta_{J,J,J}}{2}$$ Equation 59

$$Z\_add_{J,J,J}=\frac{\sigma_{J,J}\cdot\sigma_{J,J}}{C_{J,J}\cdot C_{J,J}}\cdot\zeta\_add_{J,J,J}=\left(\frac{1}{3}\right)\cdot\left(\frac{1}{3}\right)\cdot\left[\frac{-3}{20}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))}\right]$$

$$Z\_add_{J,J,J}=\frac{-1}{60}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))}=\frac{-Z_{J,J,J}}{2}$$ Equation 60

Q (probe), P (pump)

$$\zeta\_add_{J,J-1,J}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}\right]\cdot\left[\frac{M\cdot M}{J\cdot(J+1)}-\frac{1}{2}\cdot\frac{\left(J^2-M^2+J\right)}{(J\cdot(J+1))}\right]}{\sum_{M=-J}^{J}\frac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}\frac{M\cdot M}{J\cdot(J+1)}}$$

$$\zeta\_add_{J,J-1,J}=\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}=\frac{-\zeta_{J,J-1,J}}{2}$$ Equation 61

$$Z\_add_{J,J-1,J}=\frac{\sigma_{J,J-1}\cdot\sigma_{J,J}}{C_{J,J-1}\cdot C_{J,J}}\cdot\zeta\_add_{J,J-1,J}=\left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}\right]$$

$$Z\_add_{J,J-1,J}=\frac{1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J}=\frac{-Z_{J,J-1,J}}{2}$$ Equation 62

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta\_add_{J,J+1,J-1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J+M+2)}{(2\cdot J+1)\cdot(2\cdot J+2)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J+1,J-1}=\frac{-3}{20}=\frac{-\zeta_{J,J+1,J-1}}{2}$$

Equation 63

$$Z\_add_{J,J+1,J-1}=\frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J,J-1}=\left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left(\frac{-3}{20}\right)$$

$$Z\_add_{J,J+1,J-1}=\frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J,J-1}=\frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}$$

$$Z\_add_{J,J,J-1}=\frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}=\frac{-Z_{J,J,J-1}}{2}$$

Equation 64

P (probe), Q (pump)

$$\zeta\_add_{J,J,J-1}=(2\cdot J+1)\cdot\frac{\displaystyle\sum_{M=-J}^{J}\left[\frac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}\right]\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{\left(J^2+M^2-J\right)}{(J\cdot(2\cdot J+1))}\right]\right]}{\displaystyle\sum_{M=-J}^{J}\frac{(J+M+1)\cdot(J-M)}{2\cdot J\cdot(J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J,J-1}=\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}=\frac{-\zeta_{J,J,J-1}}{2}$$

Equation 65

$$Z\_add_{J,J,J-1}=\frac{\sigma_{J,J}\cdot\sigma_{J,J-1}}{C_{J,J}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J,J-1}=\left(\frac{1}{3}\right)\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{3}{20}\cdot\frac{(2\cdot J+3)}{J}\right]$$

$$Z\_add_{J,J,J-1}=\frac{1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J}=\frac{-Z_{J,J,J-1}}{2}$$

Equation 66

P (probe), P (pump)

$$\zeta\_add_{J,J-1,J-1}=(2\cdot J+1)\cdot\frac{\sum_{M=-J}^{J}\left[\frac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}\right]\cdot\left[(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}-\frac{1}{2}\cdot\left[\frac{(J^2+M^2-J)}{(J\cdot(2\cdot J+1))}\right]\right]}{\sum_{M=-J}^{J}\frac{(J-M-1)\cdot(J-M)}{2\cdot J\cdot(2\cdot J+1)}\cdot\sum_{M=-J}^{J}(-1)^2\cdot\frac{J^2-M^2}{J\cdot(2\cdot J+1)}}$$

$$\zeta\_add_{J,J-1,J-1}=\frac{-3}{20}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)}=\frac{-\zeta\_add_{J,J-1,J-1}}{2}$$ Equation 67

$$Z\_add_{J,J-1,J-1}=\frac{\sigma_{J,J-1}\cdot\sigma_{J,J-1}}{C_{J,J-1}\cdot C_{J,J-1}}\cdot\zeta\_add_{J,J-1,J-1}$$

$$Z\_add_{J,J-1,J-1}=\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{1}{3}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\right]\cdot\left[\frac{-3}{20}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)}\right]$$

$$Z\_add_{J,J-1,J-1}=\frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3)\cdot\frac{(J+1)}{J}=\frac{-Z_{J,J-1,J-1}}{2}$$ Equation 68

Summary of the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ Functions

We have summarised the $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions below. The functions are subject to the previously stated condition, in the case of $j_2 = 1$, that the functions are zero for:

- P transitions of pump and probe beams for J = ½
- P and Q transitions of pump and probe beams for J = 0

where J is the rotational quantum number of the shared lower state of the pump and probe beam transitions.

Note that we have used the convention that $\Delta\alpha_{right\text{-}circ} = \alpha_{right} - \alpha_{left}$ and $\Delta\alpha_{left\text{-}circ} = -\Delta\alpha_{right\text{-}circ} = \alpha_{left} - \alpha_{right}$. The $\zeta_{J,J',J''}$ and $Z_{J,J',J''}$ functions for left and right circularly polarised light are equal as defined.

For a circularly polarised pump beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

$$\zeta\_add_{J,J+1,J+1} = \frac{-3}{20}\cdot\frac{(2\cdot J-1)}{(2\cdot J+3)}\cdot\frac{J}{(J+1)} = \frac{-\zeta_{J,J+1,J+1}}{2} \quad \text{Equation 69}$$

$$Z\_add_{J,J+1,J+1} = \frac{-1}{60}\cdot(2\cdot J+3)\cdot(2\cdot J-1)\cdot\frac{J}{\left[(2\cdot J+1)^2\cdot(J+1)\right]} = \frac{-Z_{J,J+1,J+1}}{2} \quad \text{Equation 70}$$

R (probe), Q (pump)

$$\zeta\_add_{J,J,J+1} = \frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-\zeta_{J,J,J+1}}{2} \quad \text{Equation 71}$$

$$Z\_add_{J,J,J+1} = \frac{1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-Z_{J,J,J+1}}{2} \quad \text{Equation 72}$$

R (probe), P (pump)

$$\zeta\_add_{J,J-1,J+1} = \frac{-3}{20} = \frac{-\zeta_{J,J-1,J+1}}{2} \quad \text{Equation 73}$$

$$Z\_add_{J,J-1,J+1} = \frac{-1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)^2}\cdot(2\cdot J+3) = \frac{-Z_{J,J-1,J+1}}{2} \quad \text{Equation 74}$$

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta\_add_{J,J+1,J} = \frac{3}{20}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-\zeta_{J,J+1,J}}{2}$$ Equation 75

$$Z\_add_{J,J+1,J} = \frac{1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)}\cdot\frac{(2\cdot J-1)}{(J+1)} = \frac{-Z_{J,J+1,J}}{2}$$ Equation 76

Q probe , Q pump

$$\zeta\_add_{J,J,J} = \frac{-3}{20}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))} = \frac{-\zeta_{J,J,J}}{2}$$ Equation 77

$$Z\_add_{J,J,J} = \frac{-1}{60}\cdot\frac{\left(4\cdot J^2+4\cdot J-3\right)}{(J\cdot(J+1))} = \frac{-Z_{J,J,J}}{2}$$ Equation 78

Q (probe), P (pump)

$$\zeta\_add_{J,J-1,J} = \frac{3}{20}\cdot\frac{(2\cdot J+3)}{J} = \frac{-\zeta_{J,J-1,J}}{2}$$ Equation 79

$$Z\_add_{J,J-1,J} = \frac{1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J} = \frac{-Z_{J,J-1,J}}{2}$$ Equation 80

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta\_add_{J,J+1,J-1} = \frac{-3}{20} = \frac{-\zeta_{J,J+1,J-1}}{2}$$ Equation 81

$$Z\_add_{J,J+1,J-1} = \frac{-1}{60}\cdot\frac{(2\cdot J+3)}{(2\cdot J+1)^2}\cdot(2\cdot J-1) = \frac{-Z_{J,J+1,J-1}}{2}$$ Equation 82

P (probe), Q (pump)

$$\zeta\_add_{J,J,J-1} = \frac{3}{20}\cdot\frac{(2\cdot J+3)}{J} = \frac{-\zeta_{J,J,J-1}}{2}$$ Equation 83

$$Z\_add_{J,J,J-1} = \frac{1}{60}\cdot\frac{(2\cdot J-1)}{(2\cdot J+1)}\cdot\frac{(2\cdot J+3)}{J} = \frac{-Z_{J,J,J-1}}{2}$$ Equation 84

P (probe), P (pump)

$$\zeta\_add_{J,J-1,J-1} = \frac{-3}{20}\cdot\frac{((2\cdot J+3)\cdot(J+1))}{((2\cdot J-1)\cdot J)} = \frac{-\zeta_{J,J-1,J-1}}{2}$$ Equation 85

$$Z\_add_{J,J-1,J-1} = \frac{-1}{60} \cdot \frac{(2 \cdot J - 1)}{(2 \cdot J + 1)^2} \cdot (2 \cdot J + 3) \cdot \frac{(J+1)}{J} = \frac{-Z_{J,J-1,J-1}}{2}$$ Equation 86

For a linearly polarised pump beam

R Transitions of the probe beam, R,Q,P transitions of the pump beam

R (probe), R (pump)

$$\zeta\_add_{J,J+1,J+1} = 0$$ Equation 87

$$Z\_add_{J,J+1,J+1} = 0$$ Equation 88

R (probe), Q (pump)

$$\zeta\_add_{J,J,J+1} = 0$$ Equation 89

$$Z\_add_{J,J,J+1} = 0$$ Equation 90

R (probe), P (pump)

$$\zeta\_add_{J,J-1,J+1} = 0$$ Equation 91

$$Z\_add_{J,J-1,J+1} = 0$$ Equation 92

Q Transitions of the probe beam, R,Q,P transitions of the pump beam

Q (probe), R (pump)

$$\zeta\_add_{J,J+1,J} = 0$$ Equation 93

$$Z\_add_{J,J+1,J} = 0$$ Equation 94

Q (probe), Q (pump)

$$\zeta\_add_{J,J,J} = 0$$ Equation 95

$$Z\_add_{J,J,J} = 0$$ Equation 96

Q (probe), P (pump)

$$\zeta\_add_{J,J-1,J} = 0$$ Equation 97

$$Z\_add_{J,J-1,J} = 0$$ Equation 98

P Transitions of the probe beam, R,Q,P transitions of the pump beam

P (probe), R (pump)

$$\zeta\_add_{J,J+1,J-1} = 0$$ Equation 99

$Z\_add_{J,J+1,J-1}=0$ Equation 100

P (probe), Q (pump)

$\zeta\_add_{J,J,J-1}=0$ Equation 101

$Z_{J,J,J-1}=0$ Equation 102

P (probe), P (pump)

$\zeta\_add_{J,J-1,J-1}=0$ Equation 103

$Z\_add_{J,J-1,J-1}=0$ Equation 104

In summary, a linearly polarised pump beam will not induce circular dichroism and birefringence. However, a circularly polarised pump beam can induce linear dichroism and birefringence of opposite sign and half the magnitude of the induced linear dichroism and birefringence due to a linearly polarised pump beam.

References:

[A1] Zare, R.N., Angular Momentum. Understanding Spatial Aspects in Chemistry and Physics., John Wiley and Sons, Inc., New York, 1st Ed., 1988.

Appendix VIII: Derivation between equations [57] and [59] of Chapter VIII

Working from equation [57] in Chapter VIII,

$$\frac{E_{probe\ldots+transmitted\_\Lambda}}{E_0\cdot e^{-\alpha_{av}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}}=\left[\frac{E_{probe\_y}+i\cdot\left(\frac{a-b}{\sigma}\right)\cdot E_{probe\_x}}{2\cdot b\cdot\cos(\chi)}\right]\cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda}\cdot e^{i\cdot\frac{\Delta k}{2}\cdot\Lambda}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\\ (a+b)\cdot\cos(\chi)\\ -(a+b)\cdot\sin(\chi)\end{bmatrix}\ldots$$
$$+-\left[\left[\frac{E_{probe\_y}+i\cdot\left(\frac{a+b}{\sigma}\right)\cdot E_{probe\_x}}{2\cdot b\cdot\cos(\chi)}\right]\cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda}\cdot e^{-i\cdot\frac{\Delta k}{2}\cdot\Lambda}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\\ (a-b)\cdot\cos(\chi)\\ -(a-b)\cdot\sin(\chi)\end{bmatrix}\right]$$

Equation 1

$$\frac{E_{probe\ldots+transmitted\_\Lambda}}{E_0\cdot e^{-\alpha_{av}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}}=\frac{\left[E_{probe\_y}+i\cdot\left(\frac{a-b}{\sigma}\right)\cdot E_{probe\_x}\right]\cdot e^{-\frac{\Delta\alpha}{4}\cdot\Lambda}\cdot e^{i\cdot\frac{\Delta k}{2}\cdot\Lambda}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\\ (a+b)\cdot\cos(\chi)\\ -(a+b)\cdot\sin(\chi)\end{bmatrix}\ldots+-\left[E_{probe\_y}+i\cdot\left(\frac{a+b}{\sigma}\right)\cdot E_{probe\_x}\right]\cdot e^{\frac{\Delta\alpha}{4}\cdot\Lambda}\cdot e^{-i\cdot\frac{\Delta k}{2}\cdot\Lambda}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\\ (a-b)\cdot\cos(\chi)\\ -(a-b)\cdot\sin(\chi)\end{bmatrix}}{2\cdot b\cdot\cos(\chi)}$$

Equation 2

Writing

$$e^{i\cdot\phi}=e^{i\cdot\left(\frac{\Delta k}{2}+i\cdot\frac{\Delta\alpha}{4}\right)\cdot\Lambda}$$

Equation 3

equation [2] becomes

$$\frac{E_{probe\ldots+transmitted\_\Lambda}}{E_0\cdot e^{-\alpha_{av}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}}=\frac{\left[E_{probe\_y}+i\cdot\left(\frac{a-b}{\sigma}\right)\cdot E_{probe\_x}\right]\cdot e^{i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\\ (a+b)\cdot\cos(\chi)\\ -(a+b)\cdot\sin(\chi)\end{bmatrix}\ldots+-\left[\left[E_{probe\_y}+i\cdot\left(\frac{a+b}{\sigma}\right)\cdot E_{probe\_x}\right]\cdot e^{-i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\\ (a-b)\cdot\cos(\chi)\\ -(a-b)\cdot\sin(\chi)\end{bmatrix}\right]}{2\cdot b\cdot\cos(\chi)}$$

Equation 4

$$\frac{E_{probe} + transmitted\_\Lambda}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\left[e^{i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a+b)\cdot\cos(\chi) \\ -(a+b)\cdot\sin(\chi) \end{bmatrix} - \left[e^{-i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a-b)\cdot\cos(\chi) \\ -(a-b)\cdot\sin(\chi) \end{bmatrix} \right] \right]\cdot E_{probe\_y} \ldots + \left[(a-b)\cdot e^{i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a+b)\cdot\cos(\chi) \\ -(a+b)\cdot\sin(\chi) \end{bmatrix} \ldots + -\left[(a+b)\cdot e^{-i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a-b)\cdot\cos(\chi) \\ -(a-b)\cdot\sin(\chi) \end{bmatrix} \right] \right]\cdot E_{probe\_x}\cdot\frac{i}{\sigma}}{2\cdot b\cdot\cos(\chi)}$$

Equation 5

$$\frac{E_{probe} + transmitted\_\Lambda}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\left[e^{i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a+b)\cdot\cos(\chi) \\ -(a+b)\cdot\sin(\chi) \end{bmatrix} - e^{-i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a-b)\cdot\cos(\chi) \\ -(a-b)\cdot\sin(\chi) \end{bmatrix} \right]\cdot E_{probe\_y} \ldots + \left[e^{i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a+b)\cdot\cos(\chi) \\ -(a+b)\cdot\sin(\chi) \end{bmatrix} - e^{-i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a-b)\cdot\cos(\chi) \\ -(a-b)\cdot\sin(\chi) \end{bmatrix} \right]\cdot E_{probe\_x}\cdot\frac{i}{\sigma}\cdot a \ldots + \left[e^{i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a+b)\cdot\cos(\chi) \\ -(a+b)\cdot\sin(\chi) \end{bmatrix} + e^{-i\cdot\phi}\cdot\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi) \\ (a-b)\cdot\cos(\chi) \\ -(a-b)\cdot\sin(\chi) \end{bmatrix} \right]\cdot E_{probe\_x}\cdot\frac{-i}{\sigma}\cdot b}{2\cdot b\cdot\cos(\chi)}$$

Equation 6

$$\frac{E_{probe} + transmitted\_\Lambda}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)) \\ \cos(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi))\cdot a \ldots + \cos(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi))\cdot b \\ (-\sin(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)))\cdot a \ldots + (-\sin(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi)))\cdot b \end{bmatrix}\cdot E_{probe\_y} \ldots + \begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)) \\ (\cos(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)))\cdot a \ldots + (\cos(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi)))\cdot b \\ (-\sin(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)))\cdot a \ldots + (-\sin(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi)))\cdot b \end{bmatrix}\cdot E_{probe\_x}\cdot\frac{i}{\sigma}\cdot a \ldots + \begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi)) \\ (\cos(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi)))\cdot a \ldots + (\cos(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)))\cdot b \\ (-\sin(\chi)\cdot(\exp(i\cdot\phi) + \exp(-i\cdot\phi)))\cdot a \ldots + (-\sin(\chi)\cdot(\exp(i\cdot\phi) - \exp(-i\cdot\phi)))\cdot b \end{bmatrix}\cdot E_{probe\_x}\cdot\frac{i}{\sigma}\cdot(-b)}{2\cdot b\cdot\cos(\chi)}$$

Equation 7

$$\frac{E_{probe\ \ldots + transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\cdot(2i\cdot\sin(\phi)) \\ \cos(\chi)\cdot(2i\cdot\sin(\phi))\cdot a + \cos(\chi)\cdot(2\cdot\cos(\phi))\cdot b \\ (-\sin(\chi)\cdot(2i\cdot\sin(\phi)))\cdot a + (-\sin(\chi)\cdot(2\cdot\cos(\phi)))\cdot b \end{bmatrix}\cdot E_{probe\_y} \ldots + \begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\cdot(2i\cdot\sin(\phi)) \\ (\cos(\chi)\cdot(2i\cdot\sin(\phi)))\cdot a + (\cos(\chi)\cdot(2\cdot\cos(\phi)))\cdot b \\ (-\sin(\chi)\cdot(2i\cdot\sin(\phi)))\cdot a + (-\sin(\chi)\cdot(2\cdot\cos(\phi)))\cdot b \end{bmatrix}\cdot E_{probe\_x}\cdot\frac{i}{\sigma}\cdot a \ldots + \begin{bmatrix} i\cdot\sigma\cdot\cos(\chi)\cdot(2\cdot\cos(\phi)) \\ (\cos(\chi)\cdot(2\cdot\cos(\phi)))\cdot a + (\cos(\chi)\cdot(2i\cdot\sin(\phi)))\cdot b \\ (-\sin(\chi)\cdot(2\cdot\cos(\phi)))\cdot a + (-\sin(\chi)\cdot(2i\cdot\sin(\phi)))\cdot b \end{bmatrix}\cdot E_{probe\_x}\cdot\frac{-i}{\sigma}\cdot b}{2\cdot b\cdot\cos(\chi)}$$

Equation 8

$$\frac{E_{probe\ \ldots + transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} -2\cdot\sigma\cdot\cos(\chi)\cdot\sin(\phi) \\ 2\cdot\cos(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -2\cdot\sin(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \end{bmatrix}\cdot E_{probe\_y} \ldots + \left[\begin{bmatrix} -2\cdot\sigma\cdot\cos(\chi)\cdot\sin(\phi) \\ 2\cdot\cos(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -2\cdot\sin(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \end{bmatrix}\cdot E_{probe\_x}\cdot\frac{i}{\sigma}\cdot a \ldots + \begin{bmatrix} 2i\cdot\sigma\cdot\cos(\chi)\cdot\cos(\phi) \\ 2\cdot\cos(\chi)\cdot(\cos(\phi)\cdot a + i\cdot b\cdot\sin(\phi)) \\ -2\cdot\sin(\chi)\cdot(\cos(\phi)\cdot a + i\cdot b\cdot\sin(\phi)) \end{bmatrix}\cdot E_{probe\_x}\cdot\frac{i}{\sigma}\cdot(-b)\right]}{2\cdot b\cdot\cos(\chi)}$$

Equation 9

$$\frac{E_{probe\ \ldots + transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} -2\cdot\sigma\cdot\cos(\chi)\cdot\sin(\phi) \\ 2\cdot\cos(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -2\cdot\sin(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \end{bmatrix}\cdot E_{probe\_y} \ldots + \begin{bmatrix} 2\cdot\cos(\chi)\cdot E_{probe\_x}\cdot(-i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -2\cdot\cos(\chi)\cdot E_{probe\_x}\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \\ 2\cdot\sin(\chi)\cdot E_{probe\_x}\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \end{bmatrix}}{2\cdot b\cdot\cos(\chi)}$$

Equation 10

$$\frac{E_{probe \ldots + transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} -1\cdot\sigma\cdot\cos(\chi)\cdot\sin(\phi) \\ \cos(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -1\cdot\sin(\chi)\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \end{bmatrix}\cdot E_{probe\_y} \ldots + \begin{bmatrix} \cos(\chi)\cdot(-i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -1\cdot\cos(\chi)\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \\ \sin(\chi)\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \end{bmatrix}\cdot E_{probe\_x}}{b\cdot\cos(\chi)} \qquad \text{Equation 11}$$

or

$$\frac{E_{probe \ldots + transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} -1\cdot\sigma\cdot\sin(\phi) \\ (i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -1\cdot\frac{\sin(\chi)}{\cos(\chi)}\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \end{bmatrix}\cdot E_{probe\_y} \ldots + \begin{bmatrix} (-i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -1\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \\ \frac{\sin(\chi)}{\cos(\chi)}\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \end{bmatrix}\cdot E_{probe\_x}}{b} \qquad \text{Equation 12}$$

$$\frac{E_{probe \ldots + transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} -\sigma\cdot\sin(\phi)\cdot E_{probe\_y} \\ (i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi))\cdot E_{probe\_y} \\ \frac{-\sin(\chi)}{\cos(\chi)}\cdot(i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi))\cdot E_{probe\_y} \end{bmatrix} \ldots + \begin{bmatrix} (-i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi)) \\ -1\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \\ \frac{\sin(\chi)}{\cos(\chi)}\cdot\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma} \end{bmatrix}\cdot E_{probe\_x}}{b} \qquad \text{Equation 13}$$

$$\frac{E_{probe\_+transmitted\_\Lambda}}{E_0 \cdot e^{-\alpha_{av}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \frac{\begin{bmatrix} (-i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi))\cdot E_{probe\_x} + -\sigma\cdot\sin(\phi)\cdot E_{probe\_y} \\ -\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma}\cdot E_{probe\_x} + (i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi))\cdot E_{probe\_y} \\ -\frac{\sin(\chi)}{\cos(\chi)}\cdot\left[-\sin(\phi)\cdot(a-b)\cdot\frac{(a+b)}{\sigma}\cdot E_{probe\_x} + (i\cdot\sin(\phi)\cdot a + b\cdot\cos(\phi))\cdot E_{probe\_y}\right] \end{bmatrix}}{b} \qquad \text{Equation 14}$$

Appendix IX: Simplification of Combined Matrices

The combined Jones matrix

$$A=\begin{bmatrix}\cos(\eta) & -\sin(\eta)\\ \sin(\eta) & \cos(\eta)\end{bmatrix}\cdot\begin{bmatrix}a & b\\ c & d\end{bmatrix}\cdot\begin{bmatrix}\cos(-\eta) & -\sin(-\eta)\\ \sin(-\eta) & \cos(-\eta)\end{bmatrix}$$ Equation 1

$$A=\begin{bmatrix}\cos(\eta) & -\sin(\eta)\\ \sin(\eta) & \cos(\eta)\end{bmatrix}\cdot\begin{bmatrix}a & b\\ c & d\end{bmatrix}\cdot\begin{bmatrix}\cos(\eta) & \sin(\eta)\\ -\sin(\eta) & \cos(\eta)\end{bmatrix}$$ Equation 2

may be simplified as shown below, into a series of simple 2 x 2 matrices.

$$A=\begin{bmatrix}\cos(\eta) & -\sin(\eta)\\ \sin(\eta) & \cos(\eta)\end{bmatrix}\cdot\begin{bmatrix}a & 0\\ 0 & d\end{bmatrix}\cdot\begin{bmatrix}\cos(\eta) & \sin(\eta)\\ -\sin(\eta) & \cos(\eta)\end{bmatrix}\ldots$$
$$+\begin{bmatrix}\cos(\eta) & -\sin(\eta)\\ \sin(\eta) & \cos(\eta)\end{bmatrix}\cdot\begin{bmatrix}0 & b\\ c & 0\end{bmatrix}\cdot\begin{bmatrix}\cos(\eta) & \sin(\eta)\\ -\sin(\eta) & \cos(\eta)\end{bmatrix}$$ Equation 3

$$A=\begin{bmatrix}\cos(\eta)^2\cdot a+d-\cos(\eta)^2\cdot d & \cos(\eta)\cdot a\cdot\sin(\eta)-\sin(\eta)\cdot d\cdot\cos(\eta)\\ \cos(\eta)\cdot a\cdot\sin(\eta)-\sin(\eta)\cdot d\cdot\cos(\eta) & a-\cos(\eta)^2\cdot a+\cos(\eta)^2\cdot d\end{bmatrix}\ldots$$
$$+\begin{bmatrix}-\sin(\eta)\cdot c\cdot\cos(\eta)-\cos(\eta)\cdot b\cdot\sin(\eta) & -c+\cos(\eta)^2\cdot c+\cos(\eta)^2\cdot b\\ \cos(\eta)^2\cdot c-b+\cos(\eta)^2\cdot b & \sin(\eta)\cdot c\cdot\cos(\eta)+\cos(\eta)\cdot b\cdot\sin(\eta)\end{bmatrix}$$ Equation 4

$$A=\begin{bmatrix}\cos(\eta)^2\cdot(a-d)+d & \cos(\eta)\cdot\sin(\eta)\cdot(a-d)\\ \cos(\eta)\cdot\sin(\eta)\cdot(a-d) & a-\cos(\eta)^2\cdot(a-d)\end{bmatrix}\ldots$$
$$+\begin{bmatrix}-\cos(\eta)\cdot\sin(\eta)\cdot(c+b) & -c+\cos(\eta)^2\cdot(c+b)\\ \cos(\eta)^2\cdot(c+b)-b & \cos(\eta)\cdot\sin(\eta)\cdot(c+b)\end{bmatrix}$$ Equation 5

$$A\cdot 2=\begin{bmatrix}(1+\cos(2\cdot\eta))\cdot(a-d)+d\cdot 2 & \sin(2\cdot\eta)\cdot(a-d)\\ \sin(2\cdot\eta)\cdot(a-d) & a\cdot 2-(1+\cos(2\cdot\eta))\cdot(a-d)\end{bmatrix}\ldots$$
$$+\begin{bmatrix}-\sin(2\cdot\eta)\cdot(c+b) & -c\cdot 2+(1+\cos(2\cdot\eta))\cdot(c+b)\\ (1+\cos(2\cdot\eta))\cdot(c+b)-b\cdot 2 & \sin(2\cdot\eta)\cdot(c+b)\end{bmatrix}$$ Equation 6

Letting $p = 2\eta$

$$A\cdot 2=\begin{bmatrix}(1+\cos(p))\cdot(a-d)+d\cdot 2 & \sin(p)\cdot(a-d)\\ \sin(p)\cdot(a-d) & a\cdot 2-(1+\cos(p))\cdot(a-d)\end{bmatrix}\ldots$$
$$+\begin{bmatrix}-\sin(p)\cdot(c+b) & -c\cdot 2+(1+\cos(p))\cdot(c+b)\\ (1+\cos(p))\cdot(c+b)-b\cdot 2 & \sin(p)\cdot(c+b)\end{bmatrix}$$ Equation 7

$$A\cdot 2=\begin{bmatrix}\cos(p) & \sin(p)\\ \sin(p) & -\cos(p)\end{bmatrix}\cdot(a-d)+\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot(a+d)\ldots$$
$$+\begin{bmatrix}-\sin(p) & \cos(p)\\ \cos(p) & \sin(p)\end{bmatrix}\cdot(c+b)+\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot(c-b)$$

$$A = \frac{\begin{bmatrix} \cos(2\cdot\eta) & \sin(2\cdot\eta) \\ \sin(2\cdot\eta) & -\cos(2\cdot\eta) \end{bmatrix}\cdot(a-d) + \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot(a+d) \ldots + \begin{bmatrix} -\sin(2\cdot\eta) & \cos(2\cdot\eta) \\ \cos(2\cdot\eta) & \sin(2\cdot\eta) \end{bmatrix}\cdot(c+b) + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix}\cdot(c-b)}{2}$$ Equation 8

$$A_{c\_b\_zero} = \frac{\begin{bmatrix} \cos(2\cdot\eta) & \sin(2\cdot\eta) \\ \sin(2\cdot\eta) & -\cos(2\cdot\eta) \end{bmatrix}\cdot(a-d) + \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot(a+d)}{2}$$ Equation 9

Three cases are of interest.

Case 1: $|\eta| << 1$

For $|\eta| << 1$, the combined matrix may be approximated by

$$A(small\_\eta) = \frac{\begin{bmatrix} 1 & 2\cdot\eta \\ 2\cdot\eta & -1 \end{bmatrix}\cdot(a-d) + \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot(a+d) \ldots + \begin{bmatrix} -2\cdot\eta & 1 \\ 1 & 2\cdot\eta \end{bmatrix}\cdot(c+b) + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix}\cdot(c-b)}{2}$$ Equation 10

$$A(small\_\eta) = \begin{bmatrix} a & b \\ c & d \end{bmatrix} + \begin{bmatrix} -(c+b) & a-d \\ a-d & c+b \end{bmatrix}\cdot\eta$$ Equation 11

which for b = c = 0 becomes

$$A(small\_\eta)_{c\_b\_zero} = \begin{bmatrix} a & 0 \\ 0 & d \end{bmatrix} + \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot(a-d)\cdot\eta$$ Equation 12

Case 2: $\eta = \pi/4$

The combined matrix is given by

$$A\left(\frac{\pi}{4}\right) = \frac{\begin{bmatrix} \cos\left(2\cdot\frac{\pi}{4}\right) & \sin\left(2\cdot\frac{\pi}{4}\right) \\ \sin\left(2\cdot\frac{\pi}{4}\right) & -\cos\left(2\cdot\frac{\pi}{4}\right) \end{bmatrix}\cdot(a-d) + \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot(a+d) \ldots + \begin{bmatrix} -\sin\left(2\cdot\frac{\pi}{4}\right) & \cos\left(2\cdot\frac{\pi}{4}\right) \\ \cos\left(2\cdot\frac{\pi}{4}\right) & \sin\left(2\cdot\frac{\pi}{4}\right) \end{bmatrix}\cdot(c+b) + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix}\cdot(c-b)}{2}$$ Equation 13

$$A\left(\frac{\pi}{4}\right)=\frac{\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot(a-d)+\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot(a+d)\ldots+\begin{bmatrix}-1 & 0\\ 0 & 1\end{bmatrix}\cdot(c+b)+\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot(c-b)}{2}$$ Equation 14

$$A\left(\frac{\pi}{4}\right)=\frac{\begin{bmatrix}a+d & a-d\\ a-d & a+d\end{bmatrix}+\begin{bmatrix}-(c+b) & -(c-b)\\ c-b & c+b\end{bmatrix}}{2}$$ Equation 15

Case 3: $\eta = \pi/2$

The combined matrix is

$$A\left(\frac{\pi}{2}\right)=\frac{\begin{bmatrix}\cos\left(2\cdot\frac{\pi}{2}\right) & \sin\left(2\cdot\frac{\pi}{2}\right)\\ \sin\left(2\cdot\frac{\pi}{2}\right) & -\cos\left(2\cdot\frac{\pi}{2}\right)\end{bmatrix}\cdot(a-d)+\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot(a+d)\ldots+\begin{bmatrix}-\sin\left(2\cdot\frac{\pi}{2}\right) & \cos\left(2\cdot\frac{\pi}{2}\right)\\ \cos\left(2\cdot\frac{\pi}{2}\right) & \sin\left(2\cdot\frac{\pi}{2}\right)\end{bmatrix}\cdot(c+b)+\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot(c-b)}{2}$$ Equation 16

$$A\left(\frac{\pi}{2}\right)=\frac{\begin{bmatrix}-1 & 0\\ 0 & 1\end{bmatrix}\cdot(a-d)+\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot(a+d)\ldots+\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\cdot(c+b)+\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot(c-b)}{2}$$ Equation 17

$$A\left(\frac{\pi}{2}\right)=\begin{bmatrix}d & -c\\ -b & a\end{bmatrix}$$ Equation 18

Appendix X: Calculation of the Combined Matrices: Linear Induced Dichroism and Birefringence

Calculation of a Polariser/Birefringent-Dichroic Medium/Polariser Matrix for a Linearly Polarised Pump Beam

The combined matrix is of the form

$$C = R\left(\gamma_0 + \frac{\pi}{4}\right) \cdot F \cdot G \cdot H \cdot R\left[-\left(\gamma_0 + \frac{\pi}{4}\right)\right] \qquad \text{Equation 1}$$

where

$$F = R\left(\frac{\pi}{2} + \Delta\gamma + \Delta\theta\right) \cdot P_{imperfect} \cdot R\left[-\left(\frac{\pi}{2} + \Delta\gamma + \Delta\theta\right)\right]$$

$$F = R\left(\frac{\pi}{2}\right) \cdot R(\Delta\gamma + \Delta\theta) \cdot P_{imperfect} \cdot R(-(\Delta\gamma + \Delta\theta)) \cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 2}$$

$$G = R\left(\frac{-\pi}{4}\right) \cdot B_{linear} \cdot R\left(\frac{\pi}{4}\right) = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot R\left(\frac{-\pi}{4}\right) \cdot \begin{bmatrix} e^{-i \cdot \phi} & 0 \\ 0 & e^{i \cdot \phi} \end{bmatrix} \cdot R\left(\frac{\pi}{4}\right) \qquad \text{Equation 3}$$

$$H = R(\Delta\gamma) \cdot P_{imperfect} \cdot R(-(\Delta\gamma)) \qquad \text{Equation 4}$$

The R(ρ) matrices represent rotation through the angle, ρ, and are given by

$$R(\rho) = \begin{bmatrix} \cos(\rho) & -\sin(\rho) \\ \sin(\rho) & \cos(\rho) \end{bmatrix} \qquad \text{Equation 5}$$

while the polariser matrices are written

$$P_{imperfect} = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} \qquad \text{Equation 6}$$

Appendix IX showed that

$$A = R(\eta) \cdot \begin{bmatrix} a & b \\ c & d \end{bmatrix} \cdot R(-\eta) = \frac{\begin{bmatrix} \cos(2\cdot\eta) & \sin(2\cdot\eta) \\ \sin(2\cdot\eta) & -\cos(2\cdot\eta) \end{bmatrix} \cdot (a-d) + \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \cdot (a+d) \ldots + \begin{bmatrix} -\sin(2\cdot\eta) & \cos(2\cdot\eta) \\ \cos(2\cdot\eta) & \sin(2\cdot\eta) \end{bmatrix} \cdot (c+b) + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot (c-b)}{2} \qquad \text{Equation 7}$$

or for | η | << 1,

$$A = \begin{bmatrix} a & b \\ c & d \end{bmatrix} + \begin{bmatrix} -(c+b) & a-d \\ a-d & c+b \end{bmatrix} \cdot \eta \qquad \text{Equation 8}$$

which will be used to simplify the combined matrices in this appendix.

Using equation [8], equations [2] to [4] become

$$H=R(\Delta\gamma)\cdot P_{imperfect}\cdot R(-(\Delta\gamma))=R(\Delta\gamma)\cdot\begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix}\cdot R(-(\Delta\gamma)) \qquad \text{Equation 9}$$

$$H=\begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix}+\Delta\gamma\cdot\begin{bmatrix} -(s_1+s_2) & t_1-t_2 \\ t_1-t_2 & s_1+s_2 \end{bmatrix} \qquad \text{Equation 10}$$

$$G=R\left(\frac{-\pi}{4}\right)\cdot B_{linear}\cdot R\left(\frac{\pi}{4}\right)=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot R\left(\frac{-\pi}{4}\right)\cdot\begin{bmatrix} e^{-i\cdot\phi} & 0 \\ 0 & e^{i\cdot\phi} \end{bmatrix}\cdot R\left(\frac{\pi}{4}\right) \qquad \text{Equation 11}$$

$$G=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left[\begin{bmatrix} \cos\left(-\frac{\pi}{4}\right) & -\sin\left(-\frac{\pi}{4}\right) \\ \sin\left(-\frac{\pi}{4}\right) & \cos\left(-\frac{\pi}{4}\right) \end{bmatrix}\cdot\begin{bmatrix} e^{-i\cdot\phi} & 0 \\ 0 & e^{i\cdot\phi} \end{bmatrix}\cdot\begin{bmatrix} \cos\left(\frac{\pi}{4}\right) & -\sin\left(\frac{\pi}{4}\right) \\ \sin\left(\frac{\pi}{4}\right) & \cos\left(\frac{\pi}{4}\right) \end{bmatrix}\right] \qquad \text{Equation 12}$$

$$G=e^{-\frac{\alpha_{av}}{4}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix} \cos(\phi) & i\cdot\sin(\phi) \\ i\cdot\sin(\phi) & \cos(\phi) \end{bmatrix} \qquad \text{Equation 13}$$

where

$$\phi=\frac{\Delta k}{2}\cdot\Lambda-i\cdot\frac{\Delta\alpha}{4}\cdot\Lambda \qquad \text{Equation 14}$$

$$F=R\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\cdot P_{imperfect}\cdot R\left[-\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\right]$$

$$F=R\left(\frac{\pi}{2}\right)\cdot R(\Delta\gamma+\Delta\theta)\cdot P_{imperfect}\cdot R(-(\Delta\gamma+\Delta\theta))\cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 15}$$

$$F=R\left(\frac{\pi}{2}\right)\cdot\left[\begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix} -(s_1+s_2) & t_1-t_2 \\ t_1-t_2 & s_1+s_2 \end{bmatrix}\right]\cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 16}$$

$$F=\begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix} s_1+s_2 & -(t_1-t_2) \\ -(t_1-t_2) & -(s_1+s_2) \end{bmatrix} \qquad \text{Equation 17}$$

We remember that only zero and first order terms contribute significantly to the transmitted intensity, and, <u>assuming that</u> $t_1 >> t_2$, s_1, s_2 <u>where</u> $s_1 \sim s_2$ <u>and all small variables</u>, $\Delta\gamma$, $\Delta\theta$, t_2/t_1, s_1/t_1, s_2/t_1, <u>are the same order of magnitude</u> , these three matrix combinations reduce to

$$H=\begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix}+\Delta\gamma\cdot t_1\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \qquad \text{Equation 18}$$

$$G = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix} 1 & i\cdot\phi \\ i\cdot\phi & 1 \end{bmatrix}$$ Equation 19

$$F = \begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma + \Delta\theta)\cdot t_1\cdot\begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$ Equation 20

Letting $s_1 = s_2 = s$, $\zeta = t_2/t_1$ and $\sigma = s/t_1$ the equations above become

$$\frac{H}{t_1} = \begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$ Equation 21

$$G = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix} 1 & i\cdot\phi \\ i\cdot\phi & 1 \end{bmatrix}$$ Equation 22

$$\frac{F}{t_1} = \begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix} + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$ Equation 23

The partial combined matrix, F·G·H, assuming identical polarisers, is then given by the equation

$$\frac{F\cdot G\cdot H}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2} = \left[\begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix} \ldots + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}\right]\cdot\begin{bmatrix} 1 & i\cdot\phi \\ i\cdot\phi & 1 \end{bmatrix}\cdot\left[\begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix} \ldots + \Delta\gamma\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\right]$$ Equation 24

$$\begin{aligned}\frac{F\cdot G\cdot H}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2} = {} & \begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix}\cdot\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot\begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix} \ldots \\ & + \begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix}\cdot\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot\Delta\gamma \ldots \\ & + \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot\begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix}\cdot(\Delta\gamma + \Delta\theta) \ldots \\ & + \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot(\Delta\gamma + \Delta\theta)\cdot\Delta\gamma \ldots \\ & + \left[\begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix} \ldots \right. \\ & \quad + \begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot\Delta\gamma \ldots \\ & \quad + \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix}\cdot(\Delta\gamma + \Delta\theta) \ldots \\ & \quad \left. + \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot(\Delta\gamma + \Delta\theta)\cdot\Delta\gamma\right]\cdot i\cdot\phi\end{aligned}$$

Equation 25

$$\frac{F \cdot G \cdot H}{e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot (t_1)^2} = \left[\begin{bmatrix} \zeta - \sigma^2 & 0 \\ 0 & \zeta - \sigma^2 \end{bmatrix} \ldots + \begin{bmatrix} -\sigma & \zeta \\ 1 & -\sigma \end{bmatrix} \cdot \Delta\gamma \ldots + \begin{bmatrix} -\sigma & -\zeta \\ -1 & -\sigma \end{bmatrix} \cdot (\Delta\gamma + \Delta\theta) \ldots + \begin{bmatrix} -1 & 0 \\ 0 & -1 \end{bmatrix} \cdot (\Delta\gamma + \Delta\theta) \cdot \Delta\gamma \right] + \left[\begin{bmatrix} -\sigma + \zeta \cdot \sigma & -\sigma^2 + \zeta^2 \\ 1 - \sigma^2 & \sigma - \zeta \cdot \sigma \end{bmatrix} \ldots + \begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix} \cdot \Delta\gamma \ldots + \begin{bmatrix} -1 & -\sigma \\ -\sigma & -\zeta \end{bmatrix} \cdot (\Delta\gamma + \Delta\theta) \ldots + \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix} \cdot (\Delta\gamma + \Delta\theta) \cdot \Delta\gamma \right] \cdot i \cdot \phi$$

Equation 26

Keeping only terms to first order in the small expansion terms, this becomes

$$\frac{F \cdot G \cdot H}{e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot (t_1)^2} = \left[\begin{bmatrix} \zeta & 0 \\ 0 & \zeta \end{bmatrix} \ldots + \begin{bmatrix} 0 & 0 \\ 1 & 0 \end{bmatrix} \cdot \Delta\gamma \ldots + \begin{bmatrix} 0 & 0 \\ -1 & 0 \end{bmatrix} \cdot (\Delta\gamma + \Delta\theta) \right] + \begin{bmatrix} 0 & 0 \\ 1 & 0 \end{bmatrix} \cdot i \cdot \phi$$

Equation 27

$$\frac{F \cdot G \cdot H}{e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot (t_1)^2} = \begin{bmatrix} \zeta & 0 \\ -\Delta\theta + i \cdot \phi & \zeta \end{bmatrix}$$

Equation 28

Remembering that

$$\phi = \frac{\Delta k}{2} \cdot \Lambda - i \cdot \frac{\Delta\alpha}{4} \cdot \Lambda \quad \text{with} \quad i \cdot \phi = i \cdot \frac{\Delta k}{2} \cdot \Lambda + \frac{\Delta\alpha}{4} \cdot \Lambda$$

Equation 29

equation [28], representing the partial matrix, F·G·H, may be rewritten explicitly as

$$\frac{F \cdot G \cdot H}{e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot (t_1)^2} = \begin{bmatrix} \zeta & 0 \\ \left(\frac{\Delta\alpha}{4} \cdot \Lambda - \Delta\theta \right) + \left(i \cdot \frac{\Delta k}{2} \cdot \Lambda \right) & \zeta \end{bmatrix}$$

Equation 30

Appendix XI: Calculation of the Combined Matrices: Circular Induced Dichroism and Birefringence

Calculation of a Polariser/Birefringent-Dichroic Medium/Polariser Matrix for a Circularly Polarised Pump Beam

The combined matrix is of the form

$$C=R\left(\frac{3\,\pi}{4}\right)\cdot F\cdot G\cdot H\cdot R\left(-\frac{3\,\pi}{4}\right) \qquad \text{Equation 1}$$

where

$$F=R\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\cdot P_{imperfect}\cdot R\left[-\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\right]$$

$$F=R\left(\frac{\pi}{2}\right)\cdot R(\Delta\gamma+\Delta\theta)\cdot P_{imperfect}\cdot R(-(\Delta\gamma+\Delta\theta))\cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 2}$$

$$G=R\left(\frac{-3\cdot\pi}{4}\right)\cdot B_{circ}\cdot R\left(\frac{3\cdot\pi}{4}\right) \qquad \text{Equation 3}$$

$$G=\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{b}\cdot R\left(\frac{-3\cdot\pi}{4}\right)\cdot\begin{bmatrix} b\cdot\cos(\phi)+i\cdot a\cdot\sin(\phi) & \sigma\cdot\sin(\phi)\cdot\cos(\chi) \\ \sin(\phi)\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)} & b\cdot\cos(\phi)-i\cdot a\cdot\sin(\phi)\end{bmatrix}\cdot R\left(\frac{3\cdot\pi}{4}\right)$$

Equation 4

$$H=R\,(\Delta\gamma)\cdot P_{imperfect}\cdot R(-(\Delta\gamma)) \qquad \text{Equation 5}$$

where

$$a=\frac{\left(\Delta-\sigma^2\right)}{2}\cdot\sin(\chi)^2 \qquad \text{Equation 6}$$

or approximately

$$a=\frac{\Delta}{2}\cdot\sin(\chi)^2 \qquad \text{Equation 7}$$

and

$$b=\sqrt{\sin(\chi)^4\cdot\left(\frac{\Delta+\sigma^2}{2}\right)^2+\cos(\chi)^2\cdot\sigma^2} \qquad \text{Equation 8}$$

or approximately

$$b=\sqrt{\sin(\chi)^4\cdot\left(\frac{\Delta}{2}\right)^2+\cos(\chi)^2\cdot\sigma^2} \qquad \text{Equation 9}$$

so that

$$a^2 - b^2 = \left(\frac{\Delta - \sigma^2}{2}\right)^2 \cdot \sin(\chi)^4 - \left[\sin(\chi)^4 \cdot \left(\frac{\Delta + \sigma^2}{2}\right)^2 + \cos(\chi)^2 \cdot \sigma^2\right]$$ Equation 10

$$a^2 - b^2 = -\Delta \cdot \sigma^2 \cdot \sin(\chi)^4 - \sigma^2 \cdot \cos(\chi)^2 = -\sigma^2 \cdot \left(\cos(\chi)^2 + \Delta \cdot \sin(\chi)^4\right)$$ Equation 11

The R(ρ) matrices represent rotation through the angle, ρ,

$$R(\rho) = \begin{bmatrix} \cos(\rho) & -\sin(\rho) \\ \sin(\rho) & \cos(\rho) \end{bmatrix}$$ Equation 12

and the polariser matrices are written

$$P_{imperfect} = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix}$$ Equation 13

Appendix IX showed that

$$A = R(\eta) \cdot \begin{bmatrix} a & b \\ c & d \end{bmatrix} \cdot R(-\eta) = \frac{\begin{bmatrix} \cos(2\cdot\eta) & \sin(2\cdot\eta) \\ \sin(2\cdot\eta) & -\cos(2\cdot\eta) \end{bmatrix} \cdot (a - d) + \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \cdot (a + d) \ldots + \begin{bmatrix} -\sin(2\cdot\eta) & \cos(2\cdot\eta) \\ \cos(2\cdot\eta) & \sin(2\cdot\eta) \end{bmatrix} \cdot (c + b) + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot (c - b)}{2}$$ Equation 14

or for | η | << 1,

$$A = \begin{bmatrix} a & b \\ c & d \end{bmatrix} + \begin{bmatrix} -(c + b) & a - d \\ a - d & c + b \end{bmatrix} \cdot \eta$$ Equation 15

which will be used to simplify the combined matrices in this appendix.

$$H = R(\Delta\gamma) \cdot P_{imperfect} \cdot R(-(\Delta\gamma)) = R(\Delta\gamma) \cdot \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} \cdot R(-(\Delta\gamma))$$ Equation 16

$$H = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} + \Delta\gamma \cdot \begin{bmatrix} -(s_1 + s_2) & t_1 - t_2 \\ t_1 - t_2 & s_1 + s_2 \end{bmatrix}$$ Equation 17

$$G = \frac{\left(e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda}\right)}{b} \cdot R\left(\frac{-3\cdot\pi}{4}\right) \cdot \begin{bmatrix} b \cdot \cos(\phi) + i \cdot a \cdot \sin(\phi) & \sigma \cdot \sin(\phi) \cdot \cos(\chi) \\ \sin(\phi) \cdot \frac{(a^2 - b^2)}{\sigma \cdot \cos(\chi)} & b \cdot \cos(\phi) - i \cdot a \cdot \sin(\phi) \end{bmatrix} \cdot R\left(\frac{3\cdot\pi}{4}\right)$$

Equation 18

where

$$R\left(\frac{3\cdot\pi}{4}\right)=\frac{1}{\sqrt{2}}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}$$ Equation 19

and

$$R\left(\frac{-3\cdot\pi}{4}\right)=\frac{1}{\sqrt{2}}\cdot\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}$$ Equation 20

so that

$$G=\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{2\cdot b}\cdot\left[\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}b\cdot\cos(\phi)+i\cdot a\cdot\sin(\phi) & \sigma\cdot\sin(\phi)\cdot\cos(\chi)\\ \sin(\phi)\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)} & b\cdot\cos(\phi)-i\cdot a\cdot\sin(\phi)\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\right]$$

Equation 21

$$\frac{G}{\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{2\cdot b}}=\left[\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}b\cdot\cos(\phi)+i\cdot a\cdot\sin(\phi) & 0\\ 0 & b\cdot\cos(\phi)-i\cdot a\cdot\sin(\phi)\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\right]\ldots$$
$$+\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & \sigma\cdot\sin(\phi)\cdot\cos(\chi)\\ 0 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\ldots$$
$$+\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 0\\ \sin(\phi)\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)} & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}$$

Equation 22

$$G=\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{2\cdot b}\cdot\left[\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot(b\cdot\cos(\phi)+i\cdot a\cdot\sin(\phi))\ldots\right.$$
$$+\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot(b\cdot\cos(\phi)-i\cdot a\cdot\sin(\phi))\ldots$$
$$+\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\ 0 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot(\sigma\cdot\sin(\phi)\cdot\cos(\chi))\ldots$$
$$\left.+\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot\left[\sin(\phi)\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)}\right]\right]$$ Equation 23

$$G=\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{2\cdot b}\cdot\left[\begin{bmatrix}1&1\\1&1\end{bmatrix}\cdot(b\cdot\cos(\phi)+i\cdot a\cdot\sin(\phi))\ ...\ +\begin{bmatrix}1&-1\\-1&1\end{bmatrix}\cdot(b\cdot\cos(\phi)-i\cdot a\cdot\sin(\phi))\ ...\ +\begin{bmatrix}-1&1\\-1&1\end{bmatrix}\cdot(\sigma\cdot\sin(\phi)\cdot\cos(\chi))\ ...\ +\begin{bmatrix}-1&-1\\1&1\end{bmatrix}\cdot\left[\sin(\phi)\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)}\right]\right]$$ Equation 24

where $\phi=\frac{\Delta k}{2}\cdot\Lambda-i\cdot\frac{\Delta\alpha}{4}\cdot\Lambda$ Equation 25

$$F=R\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\cdot P_{Imperfect}\cdot R\left[-\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\right]$$

$$F=R\left(\frac{\pi}{2}\right)\cdot R(\Delta\gamma+\Delta\theta)\cdot P_{imperfect}\cdot R(-(\Delta\gamma+\Delta\theta))\cdot R\left[-\left(\frac{\pi}{2}\right)\right]$$ Equation 26

$$F=R\left(\frac{\pi}{2}\right)\cdot\left[\begin{bmatrix}t_1&s_2\\s_1&t_2\end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}-(s_1+s_2)&t_1-t_2\\t_1-t_2&s_1+s_2\end{bmatrix}\right]\cdot R\left[-\left(\frac{\pi}{2}\right)\right]$$ Equation 27

$$F=\begin{bmatrix}t_2&-s_1\\-s_2&t_1\end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}s_1+s_2&-(t_1-t_2)\\-(t_1-t_2)&-(s_1+s_2)\end{bmatrix}$$ Equation 28

We remember that only zero and first order terms contribute significantly to the transmitted intensity, and, assuming that $t_1 >> t_2$, s_1, s_2 where $s_1 \sim s_2$ and all small variables, $\Delta\gamma$, $\Delta\theta$, t_2/t_1, s_1/t_1, s_2/t_1, are the same order of magnitude , these three matrix combinations reduce to

$$H=\begin{bmatrix}t_1&s_2\\s_1&t_2\end{bmatrix}+\Delta\gamma\cdot t_1\cdot\begin{bmatrix}0&1\\1&0\end{bmatrix}$$ Equation 29

$$G=\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{2\cdot b}\cdot\left[\begin{bmatrix}1&1\\1&1\end{bmatrix}\cdot(b+i\cdot a\cdot\phi)\ ...\ +\begin{bmatrix}1&-1\\-1&1\end{bmatrix}\cdot(b-i\cdot a\cdot\phi)\ ...\ +\begin{bmatrix}-1&1\\-1&1\end{bmatrix}\cdot(\sigma\cdot\phi\cdot\cos(\chi))\ ...\ +\begin{bmatrix}-1&-1\\1&1\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)}\right]\right]$$ Equation 30

$$F=\begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma + \Delta\theta)\cdot t_1\cdot\begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$ Equation 31

Letting $s_1 = s_2 = s$, $\zeta = t_2/t_1$ and $\Sigma = s/t_1$ the equations above become

$$\frac{H}{t_1}=\begin{bmatrix} 1 & \Sigma \\ \Sigma & \zeta \end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$ Equation 32

$$G=\frac{\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}{2\cdot b}\cdot\left[\begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix}\cdot(b + i\cdot a\cdot\phi)\ \ldots + \begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix}\cdot(b - i\cdot a\cdot\phi)\ \ldots + \begin{bmatrix} -1 & 1 \\ -1 & 1 \end{bmatrix}\cdot(\sigma\cdot\phi\cdot\cos(\chi))\ \ldots + \begin{bmatrix} -1 & -1 \\ 1 & 1 \end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2 - b^2)}{\sigma\cdot\cos(\chi)}\right]\right]$$ Equation 33

$$\frac{F}{t_1}=\begin{bmatrix} \zeta & -\Sigma \\ -\Sigma & 1 \end{bmatrix} + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$ Equation 34

and the matrix, G, may be rearranged as

$$G=\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)\cdot\left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\ \ldots + \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot i\cdot\frac{a}{b}\cdot\phi\ \ldots + \begin{bmatrix} -1 & 1 \\ -1 & 1 \end{bmatrix}\cdot\left(\sigma\cdot\phi\cdot\frac{\cos(\chi)}{2\cdot b}\right)\ \ldots + \begin{bmatrix} -1 & -1 \\ 1 & 1 \end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2 - b^2)}{\sigma\cdot\cos(\chi)\cdot 2\cdot b}\right]\right]$$ Equation 35

The partial combined matrix, FGH, assuming identical polarisers, is then given by the equation

$$\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\left[\begin{bmatrix} \zeta & -\Sigma \\ -\Sigma & 1 \end{bmatrix}\ \ldots + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}\right]\cdot(S)\cdot\left[\begin{bmatrix} 1 & \Sigma \\ \Sigma & \zeta \end{bmatrix}\ \ldots + \Delta\gamma\cdot\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\right]$$ Equation 36

where $$S=\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\ \ldots + \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\cdot i\cdot\frac{a}{b}\cdot\phi\ \ldots + \begin{bmatrix} -1 & 1 \\ -1 & 1 \end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right)\ \ldots + \begin{bmatrix} -1 & -1 \\ 1 & 1 \end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2 - b^2)}{2\cdot b\cdot\sigma\cdot\cos(\chi)}\right]$$

or

$$
\begin{aligned}
\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(t_1\right)^2} = & \left[\left[\left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix} + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\right]\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot\left[\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\right]\right] \ldots \\
& + \left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix} + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\cdot i\cdot\frac{a}{b}\cdot \ldots \\
& + \left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix} \ldots + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix} \ldots + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\
& + \left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix} \ldots + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix} \ldots + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{2\cdot b\cdot\sigma\cdot\cos(\chi)}\right]
\end{aligned}
$$

Equation 37

Keeping only terms to first order in the small expansion terms, this becomes

$$
\begin{aligned}
\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(t_1\right)^2} = & \left[\left[\left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix} + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\right]\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot\left[\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\right]\right] \ldots \\
& + \begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot i\cdot\frac{a}{b}\cdot\phi \ldots \\
& + \begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\
& + \begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{2\cdot b\cdot\sigma\cdot\cos(\chi)}\right]
\end{aligned}
$$

Equation 38

$$
\begin{aligned}
\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(t_1\right)^2} = & \left[\left[\begin{matrix}\zeta+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot(\Sigma+\Delta\gamma) & \zeta\cdot(\Sigma+\Delta\gamma)+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot\zeta\\ -\Delta\theta & \zeta+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot(\Sigma+\Delta\gamma)\end{matrix}\right]\right] \ldots \\
& + \begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot i\cdot\frac{a}{b}\cdot\phi \ldots \\
& + \begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\
& + \begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{2\cdot b\cdot\sigma\cdot\cos(\chi)}\right]
\end{aligned}
$$

Equation 39

which approximates further to

$$\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta & \zeta\end{bmatrix}\ldots+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot i\cdot\frac{a}{b}\cdot\phi\ldots+\begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right)\ldots+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{2\cdot b\cdot\sigma\cdot\cos(\chi)}\right]$$ Equation 40

or

$$\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ \phi\cdot\left[\frac{(a^2-b^2)}{2\cdot b\cdot\sigma\cdot\cos(\chi)}-\frac{\sigma}{2\cdot b}\cdot\cos(\chi)\right]-\Delta\theta+i\cdot\left(\frac{a}{b}\cdot\phi\right) & \zeta\end{bmatrix}$$ Equation 41

If the intersection angle of pump and probe beams is small, the terms, a and b, may be approximated by

$$a=0$$ Equation 42

and

$$b=\sigma\cdot\cos(\chi)$$ Equation 43

and the equation above simplifies to

$$\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta-\phi & \zeta\end{bmatrix}$$ Equation 44

which is very similar to that in the case of a linearly polarised pump beam

$$\frac{(F\cdot G\cdot H)_{linear}}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta+i\cdot\phi & \zeta\end{bmatrix}$$ Equation 45

The F·G·H matrix becomes, writing the factor, ϕ, explicitly

$$\frac{(F\cdot G\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\left(\Delta\theta+\frac{\Delta k}{2}\cdot\Lambda\right)+i\cdot\frac{\Delta\alpha}{4}\cdot\Lambda & \zeta\end{bmatrix}$$ Equation 46

Appendix XII: Calculation of the Combined Matrices: Linear Induced Dichroism and Birefringence and Linearly Birefringent Inter-Polariser Optical Elements

Calculation of a Polariser/Birefringent-Dichroic Medium/Polariser Matrix for a Linearly Polarised Pump Beam with Inter-polariser Birefringent Optical Elements

Consider linearly birefringent optical elements placed between the probe beam polarisers and surrounding the region of induced dichroism and birefringence. Let the ordinary axes of the birefringent elements lie at the angles, $\pi/4 + a_i$ and $\pi/4 + a_f$, to the induced ordinary polarisation axis which lies at the angle, γ_0, to the vertical X' axis. The combined matrix may then be written as

$$C = R\left(\gamma_0 + \frac{\pi}{4}\right)\cdot F\cdot RBR_f\cdot G\cdot RBR_i\cdot H\cdot R\left[-\left(\gamma_0 + \frac{\pi}{4}\right)\right] \qquad \text{Equation 1}$$

where,

$$F = R\left(\frac{\pi}{2} + \Delta\gamma + \Delta\theta\right)\cdot P_{imperfect}\cdot R\left[-\left(\frac{\pi}{2} + \Delta\gamma + \Delta\theta\right)\right]$$

$$F = R\left(\frac{\pi}{2}\right)\cdot R(\Delta\gamma + \Delta\theta)\cdot P_{imperfect}\cdot R(-(\Delta\gamma + \Delta\theta))\cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 2}$$

$$G = R\left(\frac{-\pi}{4}\right)\cdot B_{linear}\cdot R\left(\frac{\pi}{4}\right) = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot R\left(\frac{-\pi}{4}\right)\cdot\begin{bmatrix} e^{-i\cdot\phi} & 0 \\ 0 & e^{i\cdot\phi}\end{bmatrix}\cdot R\left(\frac{\pi}{4}\right) \qquad \text{Equation 3}$$

$$G = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix} \cos\left(\frac{-\pi}{4}\right) & -\sin\left(\frac{-\pi}{4}\right) \\ \sin\left(\frac{-\pi}{4}\right) & \cos\left(\frac{-\pi}{4}\right)\end{bmatrix} R\left(\frac{-\pi}{4}\right)\cdot\begin{bmatrix} e^{-i\cdot\phi} & 0 \\ 0 & e^{i\cdot\phi}\end{bmatrix}\cdot\begin{bmatrix} \cos\left(\frac{\pi}{4}\right) & -\sin\left(\frac{\pi}{4}\right) \\ \sin\left(\frac{\pi}{4}\right) & \cos\left(\frac{\pi}{4}\right)\end{bmatrix} \qquad \text{Equation 4}$$

$$G\cdot 2 = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix} 1 & 1 \\ -1 & 1\end{bmatrix}\cdot\begin{bmatrix} e^{-i\cdot\phi} & 0 \\ 0 & e^{i\cdot\phi}\end{bmatrix}\cdot\begin{bmatrix} 1 & -1 \\ 1 & 1\end{bmatrix} \qquad \text{Equation 5}$$

$$G = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix} \cos(\phi) & i\cdot\sin(\phi) \\ i\cdot\sin(\phi) & \cos(\phi)\end{bmatrix} \qquad \text{Equation 6}$$

$$H = R(\Delta\gamma)\cdot P_{imperfect}\cdot R(-(\Delta\gamma)) \qquad \text{Equation 7}$$

and introducing the birefringent matrices via

$$B = B0 \cdot \begin{bmatrix} e^{i \cdot b} & 0 \\ 0 & e^{-i \cdot b} \end{bmatrix} = B0 \cdot \begin{bmatrix} 1 + i \cdot b & 0 \\ 0 & 1 - i \cdot b \end{bmatrix} = B0 \cdot \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} - \begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix} \cdot i \cdot b \right]$$ Equation 8

where the factor, B0, represents the effects of absorption, and is close to unity, and the exponent, b, may be complex to represent both dichroism and birefringence.

The birefringent terms may then be written

$$\frac{RBR_i}{B0} = \frac{R(a_i) \cdot B_i \cdot R(-a_i)}{B0} = \begin{bmatrix} \cos(a_i) & -\sin(a_i) \\ \sin(a_i) & \cos(a_i) \end{bmatrix} \cdot \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} - \begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix} \cdot i \cdot b_i \right] \cdot \begin{bmatrix} \cos(-a_i) & -\sin(-a_i) \\ \sin(-a_i) & \cos[(-a)_i] \end{bmatrix}$$

Equation 9

$$\frac{RBR_i}{B0} = \begin{bmatrix} \cos(a_i) & -\sin(a_i) \\ \sin(a_i) & \cos(a_i) \end{bmatrix} \cdot \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} \cos(-a_i) & -\sin[(-a)_i] \\ \sin(-a_i) & \cos(-a_i) \end{bmatrix} \cdots$$
$$+ \begin{bmatrix} \cos(a_i) & -\sin(a_i) \\ \sin(a_i) & \cos(a_i) \end{bmatrix} \cdot \begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} \cos(-a_i) & -\sin(-a_i) \\ \sin(-a_i) & \cos(-a_i) \end{bmatrix} \cdot (-i) \cdot b_i$$ Equation 10

$$\frac{RBR_i}{B0} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} -\sin(a_i)^2 + \cos(a_i)^2 & 2 \cdot \cos(a_i) \cdot \sin(a_i) \\ 2 \cdot \cos(a_i) \cdot \sin(a_i) & -\cos(a_i)^2 + \sin(a_i)^2 \end{bmatrix} \cdot (i) \cdot b_i$$ Equation 11

$$\frac{RBR_i}{B0} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} \cos(2 \cdot a_i) & \sin(2 \cdot a_i) \\ \sin(2 \cdot a_i) & -\cos(2 \cdot a_i) \end{bmatrix} \cdot i \cdot b_i$$ Equation 12

and

$$\frac{RBR_f}{B0} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} \cos(2 \cdot a_f) & \sin(2 \cdot a_f) \\ \sin(2 \cdot a_f) & -\cos(2 \cdot a_f) \end{bmatrix} \cdot i \cdot b_f$$ Equation 13

Using the results of Appendix IX for $s_1 \sim s_2$ and $t_1 >> t_2$, and for small induced dichroism and birefringence, the matrices, F, G and H may be written as

$$H = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} + \Delta\gamma \cdot t_1 \cdot \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$ Equation 14

$$G = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot \begin{bmatrix} 1 & i \cdot \phi \\ i \cdot \phi & 1 \end{bmatrix}$$ Equation 15

$$F = \begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma + \Delta\theta) \cdot t_1 \cdot \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$ Equation 16

and we may calculate the combined matrix to zero and first order in the small variables

$$g = \frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot e^{-\frac{\alpha_{av}}{2} \cdot \Lambda - i \cdot k_{av} \cdot \Lambda}}$$

Equation 17

$$g = \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \ldots + \begin{bmatrix} \cos(2 \cdot a_f) & \sin(2 \cdot a_f) \\ \sin(2 \cdot a_f) & -\cos(2 \cdot a_f) \end{bmatrix} \cdot i \cdot b_f \right] \cdot \begin{bmatrix} 1 & i \cdot \phi \\ i \cdot \phi & 1 \end{bmatrix} \cdot \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \ldots + \begin{bmatrix} \cos(2 \cdot a_f) & \sin(2 \cdot a_f) \\ \sin(2 \cdot a_f) & -\cos(2 \cdot a_f) \end{bmatrix} \cdot i \cdot b_f \right]$$

Equation 18

which, to first order in the small factors, is given by

$$\frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot \left(e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \right)} = \begin{bmatrix} 1 & i \cdot \phi \\ i \cdot \phi & 1 \end{bmatrix} \ldots + \begin{bmatrix} \cos(2 \cdot a_f) & \sin(2 \cdot a_f) \\ \sin(2 \cdot a_f) & -\cos(2 \cdot a_f) \end{bmatrix} \cdot i \cdot b_f + \begin{bmatrix} \cos(2 \cdot a_i) & \sin(2 \cdot a_i) \\ \sin(2 \cdot a_i) & -\cos(2 \cdot a_i) \end{bmatrix} \cdot i \cdot b_i$$

Equation 19

or

$$\frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot \left(e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \right)} = \begin{bmatrix} 1 & i \cdot \phi \\ i \cdot \phi & 1 \end{bmatrix} \ldots + \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \cdot i \cdot \left(b_f \cdot \cos(2 \cdot a_f) + b_i \cdot \cos(2 \cdot a_i) \right) \ldots + \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \cdot \left[i \cdot \left(b_f \cdot \sin(2 \cdot a_f) + b_i \cdot \sin(2 \cdot a_i) \right) \right]$$

Equation 20

Letting $s_1 = s_2 = s$, $\zeta = t_2/t_1$ and $\sigma = s/t_1$ the equations for F and H above become

$$\frac{H}{t_1} = \begin{bmatrix} 1 & \sigma \\ \sigma & \zeta \end{bmatrix} + \Delta\gamma \cdot \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$

Equation 21

$$\frac{F}{t_1} = \begin{bmatrix} \zeta & -\sigma \\ -\sigma & 1 \end{bmatrix} + (\Delta\gamma + \Delta\theta) \cdot \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$

Equation 22

The partial combined matrix may be written as

$$j = \frac{F \cdot \left(RBR_f \cdot G \cdot RBR_i \right) \cdot H}{B0_i \cdot B0_f \cdot (t_1)^2 \cdot L}$$

$$j=\left[\begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix}\ldots + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\left[\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\ldots + \begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}\cdot i\cdot\left(b_f\cdot\cos(2\cdot a_f)\ldots + b_i\cdot\cos(2\cdot a_i)\right)\ldots + \begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot i\cdot\left(b_f\cdot\sin(2\cdot a_f)\ldots + b_i\cdot\sin(2\cdot a_i)\right)\right]\cdot\left[\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix}\ldots + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]$$

Equation 23

where $L=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}$

Equation 24

Once again, we eliminate second order factors as they occur in the equations below

$$\frac{F\cdot\left(RBR_f\cdot G\cdot RBR_i\right)\cdot H}{B0_i\cdot B0_f\cdot(t_1)^2\cdot L}=\left[\begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\cdot\left[\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix}+\Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\ldots + \begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}\cdot\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\cos(2\cdot a_f)+b_i\cdot\cos(2\cdot a_i)\right)\right]\ldots + \begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\sin(2\cdot a_f)+b_i\cdot\sin(2\cdot a_i)\right)\right]$$

Equation 25

$$m=\frac{F\cdot\left(RBR_f\cdot G\cdot RBR_i\right)\cdot H}{B0_i\cdot B0_f\cdot(t_1)^2\cdot L}$$

$$m=\begin{bmatrix}\zeta+i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi\ldots + (i\cdot\zeta\cdot\phi-\sigma-\Delta\gamma-\Delta\theta)\cdot(\sigma+\Delta\gamma) & (\zeta+i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi)\cdot(\sigma+\Delta\gamma)\ldots + (i\cdot\zeta\cdot\phi-\sigma-\Delta\gamma-\Delta\theta)\cdot\zeta\\ -\sigma-\Delta\gamma-\Delta\theta+i\cdot\phi\ldots + (i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi+1)\cdot(\sigma+\Delta\gamma) & (-\sigma-\Delta\gamma-\Delta\theta+i\cdot\phi)\cdot(\sigma+\Delta\gamma)\ldots + (i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi+1)\cdot\zeta\end{bmatrix}\ldots + \begin{bmatrix}\zeta+\sigma^2 & 2\cdot\zeta\cdot\sigma\\ -2\cdot\sigma & -\sigma^2-\zeta\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\cos(2\cdot a_f)+b_i\cdot\cos(2\cdot a_i)\right)\right]\ldots + \begin{bmatrix}-\sigma+\zeta\cdot\sigma & -\sigma^2+\zeta^2\\ 1-\sigma^2 & \sigma-\zeta\cdot\sigma\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\sin(2\cdot a_f)+b_i\cdot\sin(2\cdot a_i)\right)\right]$$

Equation 26

which, after replacing the factor, L, becomes to first order

$$\frac{F\cdot\left(RBR_f\cdot G\cdot RBR_i\right)\cdot H}{B0_i\cdot B0_f\cdot(t_1)^2\cdot\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}=\begin{bmatrix}\zeta & 0\\ -\sigma-\Delta\gamma-\Delta\theta+i\cdot\phi+(\sigma+\Delta\gamma) & \zeta\end{bmatrix}\ldots + \begin{bmatrix}0 & 0\\ 0 & 0\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\cos(2\cdot a_f)+b_i\cdot\cos(2\cdot a_i)\right)\right]\ldots + \begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\sin(2\cdot a_f)+b_i\cdot\sin(2\cdot a_i)\right)\right]$$

Equation 27

or

$$\frac{F\cdot\left(RBR_f\cdot G\cdot RBR_i\right)\cdot H}{B0_i\cdot B0_f\cdot\left(t_1\right)^2\cdot\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta+i\cdot\phi & \zeta\end{bmatrix}+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[i\cdot\left(b_f\cdot\sin\left(2\cdot a_f\right)+b_i\cdot\sin\left(2\cdot a_i\right)\right)\right]$$

Equation 28

and in its final form reads

$$\frac{F\cdot\left(RBR_f\cdot G\cdot RBR_i\right)\cdot H}{B0_i\cdot B0_f\cdot\left(t_1\right)^2\cdot\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}=\begin{bmatrix}\zeta & 0\\ \left(\frac{\Delta\alpha}{4}\cdot\Lambda-\Delta\theta\right)+i\cdot\left(\frac{\Delta k}{2}\cdot\Lambda+b_f\cdot\sin\left(2\cdot a_f\right)+b_i\cdot\sin\left(2\cdot a_i\right)\right) & \zeta\end{bmatrix}$$

Equation 29

Appendix XIII: Calculation of the Combined Matrices: Circular Induced Dichroism and Birefringence and Linearly Birefringent Inter-Polariser Optical Elements

Calculation of a Polariser/Birefringent-Dichroic Medium/Polariser Matrix for a Circularly Polarised Pump Beam with Linearly Birefringent Inter-polariser Optical Elements

By comparison with Appendix XII, we define the ordinary axis of the two linearly birefringent matrices to lie at the angles, $\pi/4 + a_i$ and $\pi/4 + a_f$, to that of the region of induced birefringence and dichroism.

The birefringent matrices can then be written in the form

$$\frac{R\left(a+\frac{\pi}{4}\right)\cdot B\cdot R\left[-\left(a+\frac{\pi}{4}\right)\right]}{B0}=\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}+\begin{bmatrix}\cos\left[2\cdot\left(a+\frac{\pi}{4}\right)\right] & \sin\left[2\cdot\left(a+\frac{\pi}{4}\right)\right]\\ \sin\left[2\cdot\left(a+\frac{\pi}{4}\right)\right] & -\cos\left[2\cdot\left(a+\frac{\pi}{4}\right)\right]\end{bmatrix}\cdot i\cdot b \quad \text{Equation 1}$$

$$\frac{R\left(a+\frac{\pi}{4}\right)\cdot B\cdot R\left[-\left(a+\frac{\pi}{4}\right)\right]}{B0}=\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}+\begin{bmatrix}-\sin(2\cdot a) & \cos(2\cdot a)\\ \cos(2\cdot a) & \sin(2\cdot a)\end{bmatrix}\cdot i\cdot b \quad \text{Equation 2}$$

The combined matrix from Appendix XI is rewritten as

$$C=R\left(\frac{3\,\pi}{4}\right)\cdot F\cdot(BGB)\cdot H\cdot R\left(-\frac{3\,\pi}{4}\right) \quad \text{Equation 3}$$

where

$$F=R\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\cdot P_{imperfect}\cdot R\left[-\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\right]$$

$$F=R\left(\frac{\pi}{2}\right)\cdot R(\Delta\gamma+\Delta\theta)\cdot P_{imperfect}\cdot R(-(\Delta\gamma+\Delta\theta))\cdot R\left[-\left(\frac{\pi}{2}\right)\right] \quad \text{Equation 4}$$

$$BGB=R\left(\frac{-3\cdot\pi}{4}\right)\cdot\left[R\left(a_f+\frac{\pi}{4}\right)\cdot B_f\cdot R\left[-\left(a_f+\frac{\pi}{4}\right)\right]\right]\cdot B_{circ}\cdot\left[R\left(a_i+\frac{\pi}{4}\right)\cdot B_i\cdot R\left[-\left(a_i+\frac{\pi}{4}\right)\right]\right]\cdot R\left(\frac{3\cdot\pi}{4}\right) \quad \text{Equation 5}$$

where

$$B_{circ}=\frac{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}}{b}\cdot\begin{bmatrix}b\cdot\cos(\phi)\ ...\ +i\cdot a\cdot\sin(\phi) & \sigma\cdot\sin(\phi)\cdot\cos(\chi)\\ \sin(\phi)\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)} & b\cdot\cos(\phi)\ ...\ +-i\cdot a\cdot\sin(\phi)\end{bmatrix} \quad \text{Equation 6}$$

$$H=R(\Delta\gamma)\cdot P_{imperfect}\cdot R(-(\Delta\gamma)) \quad \text{Equation 7}$$

and the factors, a and b, have been defined in Appendix XII.

Appendix X showed that

$$H=\begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix} -(s_1+s_2) & t_1-t_2 \\ t_1-t_2 & s_1+s_2 \end{bmatrix}$$ Equation 8

and

$$F=\begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix} s_1+s_2 & -(t_1-t_2) \\ -(t_1-t_2) & -(s_1+s_2) \end{bmatrix}$$ Equation 9

leaving us to determine the matrix description of BGB to zero and first order.

$$\frac{BGB}{B0_i\cdot B0_f}=R\left(\frac{-3\cdot\pi}{4}\right)\cdot(J)\cdot R\left(\frac{3\cdot\pi}{4}\right)$$ Equation 10

where

$$J=\begin{bmatrix}\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots \\ +\begin{bmatrix}-\sin(2\cdot a_f) & \cos(2\cdot a_f)\\ \cos(2\cdot a_f) & \sin(2\cdot a_f)\end{bmatrix}\cdot i\cdot b_f\end{bmatrix}\cdot B_{circ}\cdot\begin{bmatrix}\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots \\ +\begin{bmatrix}-\sin(2\cdot a_i) & \cos(2\cdot a_i)\\ \cos(2\cdot a_i) & \sin(2\cdot a_i)\end{bmatrix}\cdot i\cdot b_i\end{bmatrix}$$

and the induced birefringence/dichroism component may be approximated by

$$B_{circ}=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix}1+i\cdot\frac{a}{b}\cdot\phi & \frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\\ \phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b} & 1-i\cdot\frac{a}{b}\cdot\phi\end{bmatrix}$$

$$B_{circ}=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\begin{bmatrix}\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots\\ +\begin{bmatrix}1&0\\0&-1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots\\ +\begin{bmatrix}0&1\\0&0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ldots\\ +\begin{bmatrix}0&0\\1&0\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b}\right]\end{bmatrix}$$ Equation 11

Our matrix

$$\frac{BGB}{B0_i\cdot B0_f}=R\left(\frac{-3\cdot\pi}{4}\right)\cdot(M)\cdot R\left(\frac{3\cdot\pi}{4}\right)$$

where

$$M=\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots+\begin{bmatrix}-\sin(2\cdot a_f)&\cos(2\cdot a_f)\\\cos(2\cdot a_f)&\sin(2\cdot a_f)\end{bmatrix}\cdot i\cdot b_f\right]\cdot B_{circ}\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots+\begin{bmatrix}-\sin(2\cdot a_i)&\cos(2\cdot a_i)\\\cos(2\cdot a_i)&\sin(2\cdot a_i)\end{bmatrix}\cdot i\cdot b_i\right]$$ Equation 12

is then approximated to zero and first order by

$$\frac{BGB}{L\cdot B0_i\cdot B0_f}=R\left(\frac{-3\cdot\pi}{4}\right)\cdot(N)\cdot R\left(\frac{3\cdot\pi}{4}\right)$$ Equation 13

where

$$N=\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots+\begin{bmatrix}-\sin(2\cdot a_f)&\cos(2\cdot a_f)\\\cos(2\cdot a_f)&\sin(2\cdot a_f)\end{bmatrix}\cdot i\cdot b_f\right]\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots+\begin{bmatrix}-\sin(2\cdot a_i)&\cos(2\cdot a_i)\\\cos(2\cdot a_i)&\sin(2\cdot a_i)\end{bmatrix}\cdot i\cdot b_i\right]\ldots$$
$$+\begin{bmatrix}1&0\\0&1\end{bmatrix}\cdot\begin{bmatrix}1&0\\0&-1\end{bmatrix}\cdot\begin{bmatrix}1&0\\0&1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots$$
$$+\begin{bmatrix}1&0\\0&1\end{bmatrix}\cdot\begin{bmatrix}0&1\\0&0\end{bmatrix}\cdot\begin{bmatrix}1&0\\0&1\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ldots$$
$$+\begin{bmatrix}1&0\\0&1\end{bmatrix}\cdot\begin{bmatrix}0&0\\1&0\end{bmatrix}\cdot\begin{bmatrix}1&0\\0&1\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b}\right]$$

Equation 14

and

$$L=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}$$ Equation 15

so that

$$\frac{BGB}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot B0_i\cdot B0_f}=R\left(\frac{-3\cdot\pi}{4}\right)\cdot\left[\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots+\begin{bmatrix}-\sin(2\cdot a_f)&\cos(2\cdot a_f)\\\cos(2\cdot a_f)&\sin(2\cdot a_f)\end{bmatrix}\cdot i\cdot b_f\ldots+\begin{bmatrix}-\sin(2\cdot a_i)&\cos(2\cdot a_i)\\\cos(2\cdot a_i)&\sin(2\cdot a_i)\end{bmatrix}\cdot i\cdot b_i\right]\ldots+\begin{bmatrix}1&0\\0&-1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots+\begin{bmatrix}0&1\\0&0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ldots+\begin{bmatrix}0&0\\1&0\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b}\right]\right]\cdot R\left(3\cdot\frac{\pi}{2}\right)$$

Equation 16

$$\frac{BGB\cdot 2}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(B0_i\cdot B0_f\right)}=\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\left[\left[\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\ \ldots\ +\begin{bmatrix}-\sin\left(2\cdot a_f\right) & \cos\left(2\cdot a_f\right)\\ \cos\left(2\cdot a_f\right) & \sin\left(2\cdot a_f\right)\end{bmatrix}\cdot i\cdot b_f\ \ldots\ +\begin{bmatrix}-\sin\left(2\cdot a_i\right) & \cos\left(2\cdot a_i\right)\\ \cos\left(2\cdot a_i\right) & \sin\left(2\cdot a_i\right)\end{bmatrix}\cdot i\cdot b_i\right]\ \ldots\ +\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ \ldots\ +\begin{bmatrix}0 & 1\\ 0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ \ldots\ +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\right]\ \ldots\ \cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}$$

Equation 17

$$\frac{BGB\cdot 2}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(B0_i\cdot B0_f\right)}=\left[\left[\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\ \ldots\ +\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}-\sin\left(2\cdot a_f\right) & \cos\left(2\cdot a_f\right)\\ \cos\left(2\cdot a_f\right) & \sin\left(2\cdot a_f\right)\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot i\cdot b_f\ \ldots\ +\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}-\sin\left(2\cdot a_i\right) & \cos\left(2\cdot a_i\right)\\ \cos\left(2\cdot a_i\right) & \sin\left(2\cdot a_i\right)\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot i\cdot b_i\right]\ \ldots\ +\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ \ldots\ +\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\ 0 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ \ldots\ +\begin{bmatrix}-1 & 1\\ -1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & -1\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\right]$$

$$\frac{BGB\cdot 2}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(B0_i\cdot B0_f\right)}=\left[\left[\begin{bmatrix}2 & 0\\ 0 & 2\end{bmatrix}\ \ldots\ +\begin{bmatrix}-2\cdot\cos\left(2\cdot a_f\right) & -2\cdot\sin\left(2\cdot a_f\right)\\ -2\cdot\sin\left(2\cdot a_f\right) & 2\cdot\cos\left(2\cdot a_f\right)\end{bmatrix}\cdot i\cdot b_f\ \ldots\ +\begin{bmatrix}-2\cdot\cos\left(2\cdot a_i\right) & -2\cdot\sin\left(2\cdot a_i\right)\\ -2\cdot\sin\left(2\cdot a_i\right) & 2\cdot\cos\left(2\cdot a_i\right)\end{bmatrix}\cdot i\cdot b_i\right]\ \ldots\ +\begin{bmatrix}0 & 2\\ 2 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ \ldots\ +\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ \ldots\ +\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\right]$$

Equation 18

$$\frac{BGB}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot\left(B0_i\cdot B0_f\right)}=\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\ldots$$
$$+\begin{bmatrix}-1\cdot\cos\left(2\cdot a_f\right) & 1\cdot\sin\left(2\cdot a_f\right)\\ -1\cdot\sin\left(2\cdot a_f\right) & \cos\left(2\cdot a_f\right)\end{bmatrix}\cdot i\cdot b_f\ldots$$
$$+\begin{bmatrix}-1\cdot\cos\left(2\cdot a_i\right) & 1\cdot\sin\left(2\cdot a_i\right)\\ -1\cdot\sin\left(2\cdot a_i\right) & \cos\left(2\cdot a_i\right)\end{bmatrix}\cdot i\cdot b_i\ldots$$
$$+\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots$$
$$+\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right)+\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{2\cdot\sigma\cdot\cos(\chi)\cdot b}\right]$$

Equation 19

Letting $s_1 = s_2 = s$, $\zeta = t_2/t_1$ and $\Sigma = s/t_1$ the equations for H and F above become

$$\frac{H}{t_1}=\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix}+\Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}$$

Equation 20

$$\frac{F}{t_1}=\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}$$

Equation 21

The partial combined matrix, FBGBH, assuming identical polarisers, is then given by the equation

$$\frac{(F\cdot BGB\cdot H)}{L\cdot B0_i\cdot B0_f\cdot\left(t_1\right)^2}=\frac{F}{t_1}\cdot\left[\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\ldots\right.$$
$$+\begin{bmatrix}-1\cdot\cos\left(2\cdot a_f\right) & 1\cdot\sin\left(2\cdot a_f\right)\\ -1\cdot\sin\left(2\cdot a_f\right) & \cos\left(2\cdot a_f\right)\end{bmatrix}\cdot i\cdot b_f\ldots$$
$$+\begin{bmatrix}-1\cdot\cos\left(2\cdot a_i\right) & 1\cdot\sin\left(2\cdot a_i\right)\\ -1\cdot\sin\left(2\cdot a_i\right) & \cos\left(2\cdot a_i\right)\end{bmatrix}\cdot i\cdot b_i\ldots$$
$$+\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots$$
$$+\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right)\ldots$$
$$\left.+\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2-b^2\right)}{2\cdot\sigma\cdot\cos(\chi)\cdot b}\right]\right]\cdot\frac{H}{t_1}$$

Equation 22

or

$$\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot B0_i\cdot B0_f\cdot (t_1)^2}=\left[\left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\right]\cdot\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix}+\Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\ldots$$
$$+\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1\cdot\cos(2\cdot a_f) & 1\cdot\sin(2\cdot a_f)\\ -1\cdot\sin(2\cdot a_f) & \cos(2\cdot a_f)\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot(i\cdot b_f)\ldots$$
$$+\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1\cdot\cos(2\cdot a_i) & 1\cdot\sin(2\cdot a_i)\\ -1\cdot\sin(2\cdot a_i) & \cos(2\cdot a_i)\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot(i\cdot b_i)\ldots$$
$$+\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots$$
$$+\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right)\ldots$$
$$+\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{2\cdot\sigma\cdot\cos(\chi)\cdot b}\right]$$

Equation 23

$$\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot B0_i\cdot B0_f\cdot (t_1)^2}=\begin{bmatrix}\zeta+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot(\Sigma+\Delta\gamma) & \zeta\cdot(\Sigma+\Delta\gamma)+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot\zeta\\ -\Delta\theta & \zeta+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot(\Sigma+\Delta\gamma)\end{bmatrix}\ldots$$
$$+\begin{bmatrix}0 & 0\\ -\sin(2\cdot a_f) & 0\end{bmatrix}\cdot(i\cdot b_f)\ldots$$
$$+\begin{bmatrix}0 & 0\\ -\sin(2\cdot a_i) & 0\end{bmatrix}\cdot(i\cdot b_i)\ldots$$
$$+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots$$
$$+\begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{2\cdot b}\cdot\phi\cdot\cos(\chi)\right)\ldots$$
$$+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{2\cdot\sigma\cdot\cos(\chi)\cdot b}\right]$$

Equation 24

which, on retaining only the zero and first order terms becomes

$$\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot B0_i\cdot B0_f\cdot (t_1)^2}=\begin{bmatrix}\zeta & 0\\ \left[\phi\cdot\left[\frac{(a^2-b^2)}{2\cdot\sigma\cdot\cos(\chi)\cdot b}-\frac{\sigma}{2\cdot b}\cdot\cos(\chi)\right]-\Delta\theta\ldots\atop +i\cdot\left[\frac{a}{b}\cdot\phi-\left(b_f\cdot\sin(2\cdot a_f)+b_i\cdot\sin(2\cdot a_i)\right)\right]\right] & \zeta\end{bmatrix}$$

Equation 25

If the intersection angle of pump and probe beams is small, the terms a and b may be approximated

by $a=0$ Equation 26

and $b=\sigma\cdot\cos(\chi)$ Equation 27

and the equation above simplifies to

$$\frac{(F \cdot BGB \cdot H)}{e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot B0_i \cdot B0_f \cdot (t_1)^2} = \begin{bmatrix} \zeta & 0 \\ -(\phi + \Delta\theta) \ldots + -i \cdot \left(\left(b_f \cdot \sin(2 \cdot a_f) + b_l \cdot \sin(2 \cdot a_l)\right)\right) & \zeta \end{bmatrix}$$ Equation 28

which is an extension of the no birefringent element case

$$\frac{(F \cdot G \cdot H)_{no\_birefringent\_element}}{e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot (t_1)^2} = \begin{bmatrix} \zeta & 0 \\ -\Delta\theta - \phi & \zeta \end{bmatrix}$$ Equation 29

and compares to the case of a linearly polarised pump beam

$$\frac{\left[F \cdot \left(RBR_f \cdot G \cdot RBR_i\right) \cdot H\right]_{linear \ldots + birefringent\_elements}}{B0_i \cdot B0_f \cdot (t_1)^2 \cdot \left(e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda}\right)} = \begin{bmatrix} \zeta & 0 \\ -\Delta\theta + i \cdot \left[\phi + \left(b_f \cdot \sin(2 \cdot a_f) + b_l \cdot \sin(2 \cdot a_l)\right)\right] & \zeta \end{bmatrix}$$

Equation 30

Appendix XIV: Calculation of the Combined Matrices: Linear Induced Dichroism and Birefringence and Circularly Birefringent Inter-Polariser Optical Elements

Calculation of a Polariser/Birefringent-Dichroic Medium/Polariser Matrix for a Circularly Polarised Pump Beam with Inter-polariser Birefringent Optical Elements

Consider circularly birefringent optical elements in the probe beam path placed between the probe beam polarisers and surrounding the region of induced dichroism and birefringence. Let the birefringent elements be aligned with respect to a marked axis at the angles, $\pi/4 + a_i$ and $\pi/4 + a_f$, to the induced ordinary polarisation axis which lies at the angle, γ_o, to the vertical X' axis. The combined matrix may then be written as

$$C = R\left(\gamma_0 + \frac{\pi}{4}\right) \cdot F \cdot RBR_f \cdot G \cdot RBR_i \cdot H \cdot R\left[-\left(\gamma_0 + \frac{\pi}{4}\right)\right] \qquad \text{Equation 1}$$

where,

$$F = R\left(\frac{\pi}{2} + \Delta\gamma + \Delta\theta\right) \cdot P_{imperfect} \cdot R\left[-\left(\frac{\pi}{2} + \Delta\gamma + \Delta\theta\right)\right]$$

$$F = R\left(\frac{\pi}{2}\right) \cdot R(\Delta\gamma + \Delta\theta) \cdot P_{imperfect} \cdot R(-(\Delta\gamma + \Delta\theta)) \cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 2}$$

$$G = R\left(\frac{-\pi}{4}\right) \cdot B_{linear} \cdot R\left(\frac{\pi}{4}\right) = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot R\left(\frac{-\pi}{4}\right) \cdot \begin{bmatrix} e^{-i\phi} & 0 \\ 0 & e^{i\phi} \end{bmatrix} \cdot R\left(\frac{\pi}{4}\right) \qquad \text{Equation 3}$$

$$G = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot \begin{bmatrix} \cos\left(\frac{-\pi}{4}\right) & -\sin\left(\frac{-\pi}{4}\right) \\ \sin\left(\frac{-\pi}{4}\right) & \cos\left(\frac{-\pi}{4}\right) \end{bmatrix} R\left(\frac{-\pi}{4}\right) \cdot \begin{bmatrix} e^{-i\phi} & 0 \\ 0 & e^{i\phi} \end{bmatrix} \cdot \begin{bmatrix} \cos\left(\frac{\pi}{4}\right) & -\sin\left(\frac{\pi}{4}\right) \\ \sin\left(\frac{\pi}{4}\right) & \cos\left(\frac{\pi}{4}\right) \end{bmatrix}$$

Equation 4

$$G \cdot 2 = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot \begin{bmatrix} 1 & 1 \\ -1 & 1 \end{bmatrix} \cdot \begin{bmatrix} e^{-i\phi} & 0 \\ 0 & e^{i\phi} \end{bmatrix} \cdot \begin{bmatrix} 1 & -1 \\ 1 & 1 \end{bmatrix} \qquad \text{Equation 5}$$

$$G = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot \begin{bmatrix} \cos(\phi) & i \cdot \sin(\phi) \\ i \cdot \sin(\phi) & \cos(\phi) \end{bmatrix} \qquad \text{Equation 6}$$

$$H = R(\Delta\gamma) \cdot P_{imperfect} \cdot R(-(\Delta\gamma)) \qquad \text{Equation 7}$$

and introducing the birefringent matrices via

$$B = B0 \cdot e^{-i \cdot b} \cdot \begin{bmatrix} \cos(b) & \sin(b) \\ -\sin(b) & \cos(b) \end{bmatrix} = B0 \cdot \begin{bmatrix} \cos(b) & \sin(b) \\ -\sin(b) & \cos(b) \end{bmatrix} = B0 \cdot \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix} \cdot b \right] \quad \text{Equation 8}$$

where the factor, B0, represents the effects of absorption, and is close to unity, and the exponent, b, may be complex to represent both dichroism and birefringence. Note that the introduced phase difference between the two probe beam components is 2b.

The birefringent terms may then be written

$$\frac{RBR_i}{B0} = \frac{R(a_i) \cdot B_i \cdot R(-a_i)}{B0} = \begin{bmatrix} \cos(a_i) & -\sin(a_i) \\ \sin(a_i) & \cos(a_i) \end{bmatrix} \cdot \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot b_i \right] \cdot \begin{bmatrix} \cos(-a_i) & -\sin(-a_i) \\ \sin(-a_i) & \cos(-a_i) \end{bmatrix}$$

Equation 9

$$\frac{RBR_i}{B0} = \begin{bmatrix} \cos(a_i) & -\sin(a_i) \\ \sin(a_i) & \cos(a_i) \end{bmatrix} \cdot \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} \cos(-a_i) & -\sin(-a_i) \\ \sin(-a_i) & \cos(-a_i) \end{bmatrix} \ldots$$
$$+ \begin{bmatrix} \cos(a_i) & -\sin(a_i) \\ \sin(a_i) & \cos(a_i) \end{bmatrix} \cdot \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot \begin{bmatrix} \cos(-a_i) & -\sin(-a_i) \\ \sin(-a_i) & \cos(-a_i) \end{bmatrix} \cdot b_i$$

Equation 10

$$\frac{RBR_i}{B0} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot b$$

Equation 11

indicating that the action of the circularly birefringent matrix is independent of the input probe beam polarisation.

From the results of Appendix IX, we have

$$H = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} + \Delta\gamma \cdot t_1 \cdot \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$

Equation 12

$$F = \begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma + \Delta\theta) \cdot t_1 \cdot \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$

Equation 13

and for small induced dichroism and birefringence, the matrix, G, may be written as

$$G = e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda} \cdot \begin{bmatrix} 1 & i \cdot \phi \\ i \cdot \phi & 1 \end{bmatrix}$$

Equation 14

To zero and first order, the combined matrix is then given by

$$\frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot e^{-\frac{\alpha_{av}}{2} \cdot \Lambda} \cdot e^{-i \cdot k_{av} \cdot \Lambda}} = \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot b_f \right] \cdot \begin{bmatrix} 1 & i \cdot \phi \\ i \cdot \phi & 1 \end{bmatrix} \cdot \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot b_i \right]$$

Equation 15

$$\frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot e^{-\frac{\alpha_{av}}{2}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\ldots + \begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot b_f \ldots + \begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\cdot\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot b_i$$

Equation 16

$$\frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot e^{-\frac{\alpha_{av}}{2}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix} + \begin{bmatrix}-i\cdot\phi & -1\\ 1 & i\cdot\phi\end{bmatrix}\cdot b_f + \begin{bmatrix}i\cdot\phi & -1\\ 1 & -i\cdot\phi\end{bmatrix}\cdot b_i$$

Equation 17

which, to first order in the small factors, is given by

$$\frac{RBR_f \cdot G \cdot RBR_i}{B0_i \cdot B0_f \cdot e^{-\frac{\alpha_{av}}{2}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}} = \begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\ldots + \begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot b_f + \begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot b_i = \begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix} + \begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot\left(b_f + b_i\right)$$

Equation 18

Letting $s_1 = s_2 = s$, $\zeta = t_2/t_1$ and $\sigma = s/t_1$ the equations for F and H above become

$$\frac{H}{t_1} = \begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}$$

Equation 19

$$\frac{F}{t_1} = \begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix} + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}$$

Equation 20

The partial combined matrix may be written as

$$J = \frac{F\cdot\left(RBR_f \cdot G \cdot RBR_i\right)\cdot H}{B0_i \cdot B0_f \cdot \left(t_1\right)^2 \cdot e^{-\frac{\alpha_{av}}{2}\cdot\Lambda} \cdot e^{-i\cdot k_{av}\cdot\Lambda}}$$

$$J = \left[\begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix}\ldots + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\left[\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\ldots + \begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot\left(b_f + b_i\right)\right]\cdot\left[\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix}\ldots + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]$$

Equation 21

Once again, we eliminate second order factors as they occur in the equations below

$$J = \left[\begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix} + (\Delta\gamma + \Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\begin{bmatrix}1 & i\cdot\phi\\ i\cdot\phi & 1\end{bmatrix}\cdot\left[\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\ldots + \begin{bmatrix}\zeta & -\sigma\\ -\sigma & 1\end{bmatrix}\cdot\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & \sigma\\ \sigma & \zeta\end{bmatrix}\cdot\left(b_f + b_i\right)$$

Equation 22

$$J=\begin{bmatrix} \zeta + i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi \ldots \\ +(i\cdot\zeta\cdot\phi-\sigma-\Delta\gamma-\Delta\theta)\cdot(\sigma+\Delta\gamma) & (\zeta+i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi)\cdot(\sigma+\Delta\gamma)\ldots \\ & +(i\cdot\zeta\cdot\phi-\sigma-\Delta\gamma-\Delta\theta)\cdot\zeta \\ -\sigma-\Delta\gamma-\Delta\theta+i\cdot\phi\ldots & (-\sigma-\Delta\gamma-\Delta\theta+i\cdot\phi)\cdot(\sigma+\Delta\gamma)\ldots \\ +(i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi+1)\cdot(\sigma+\Delta\gamma) & +(i\cdot(-\sigma-\Delta\gamma-\Delta\theta)\cdot\phi+1)\cdot\zeta \end{bmatrix}\ldots$$
$$+\begin{bmatrix} -\sigma-\zeta\cdot\sigma & -\sigma^2-\zeta^2 \\ 1+\sigma^2 & \sigma+\zeta\cdot\sigma \end{bmatrix}\cdot(b_f+b_i)$$

Equation 23

where $L=e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}$ Equation 24

which may be approximated to first order by

$$\frac{F\cdot(RBR_f\cdot G\cdot RBR_i)\cdot H}{B0_i\cdot B0_f\cdot(t_1)^2\cdot\left(e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\right)}=\begin{bmatrix} \zeta & 0 \\ -\Delta\theta+i\cdot\phi & \zeta \end{bmatrix}+\begin{bmatrix} 0 & 0 \\ 1 & 0 \end{bmatrix}\cdot(b_f+b_i)$$ Equation 25

Note that, in this case, there is no angular dependence associated with the birefringence factor.

Appendix XV: Calculation of the Combined Matrices: Circular Induced Dichroism and Birefringence and Circularly Birefringent Inter-Polariser Optical Elements

Calculation of a Polariser/Birefringent-Dichroic Medium/Polariser Matrix for a Circularly Polarised Pump Beam with Linearly Birefringent Inter-polariser Optical Elements

Circularly birefringent matrices were shown to be independent of rotational orientation in Appendix XIV and can be written in a form where we use the factor, β, to avoid confusion with the a and b factors below

$$\frac{B}{B0}=\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}+\begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\beta \qquad \text{Equation 1}$$

The combined matrix is then written

$$C=R\left(\frac{3\pi}{4}\right)\cdot F\cdot(BGB)\cdot H\cdot R\left(-\frac{3\pi}{4}\right) \qquad \text{Equation 2}$$

where

$$F=R\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\cdot P_{imperfect}\cdot R\left[-\left(\frac{\pi}{2}+\Delta\gamma+\Delta\theta\right)\right]$$

$$F=R\left(\frac{\pi}{2}\right)\cdot R(\Delta\gamma+\Delta\theta)\cdot P_{imperfect}\cdot R(-(\Delta\gamma+\Delta\theta))\cdot R\left[-\left(\frac{\pi}{2}\right)\right] \qquad \text{Equation 3}$$

$$BGB=R\left(\frac{-3\cdot\pi}{4}\right)\cdot B_f\cdot B_{circ}\cdot B_i\cdot R\left(\frac{3\cdot\pi}{4}\right) \qquad \text{Equation 4}$$

$$J=\frac{BGB}{B0_i\cdot B0_f\cdot L} \qquad \text{Equation 5}$$

where

$$J=R\left(\frac{-3\cdot\pi}{4}\right)\cdot\left[\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}\ldots+\begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\beta_f\right]\cdot\frac{\begin{bmatrix}b\cdot\cos(\phi)\ldots+i\cdot a\cdot\sin(\phi) & \sigma\cdot\sin(\phi)\cdot\cos(\chi)\\ \sin(\phi)\cdot\dfrac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)} & b\cdot\cos(\phi)\ldots+-i\cdot a\cdot\sin(\phi)\end{bmatrix}}{b}\cdot\left[\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}\ldots+\begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\beta_i\right]\cdot R\left(\frac{3\cdot\pi}{4}\right)$$

$$L = e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}$$ Equation 6

and

$$H = R(\Delta\gamma)\cdot P_{imperfect}\cdot R(-(\Delta\gamma))$$ Equation 7

The factors, a and b, are defined in Appendix XII

Appendix IX showed that the matrices, H and F, may be written

$$H = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} + \Delta\gamma\cdot\begin{bmatrix} -(s_1+s_2) & t_1-t_2 \\ t_1-t_2 & s_1+s_2 \end{bmatrix}$$ Equation 8

and

$$F = \begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix} s_1+s_2 & -(t_1-t_2) \\ -(t_1-t_2) & -(s_1+s_2) \end{bmatrix}$$ Equation 9

For small induced dichroism and birefringence, the B·G·B matrix may be approximated by

$$\frac{BGB}{B0_i\cdot B0_f\cdot L} = R\left(\frac{-3\cdot\pi}{4}\right)\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots + \begin{bmatrix}0&-1\\1&0\end{bmatrix}\cdot\beta_f\right]\cdot\begin{bmatrix} 1+i\cdot\frac{a}{b}\cdot\phi & \frac{\sigma}{b}\cdot\phi\cdot\cos(\chi) \\ \phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b} & 1-i\cdot\frac{a}{b}\cdot\phi \end{bmatrix}\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots + \begin{bmatrix}0&-1\\1&0\end{bmatrix}\cdot\beta_i\right]\cdot R\left(\frac{3\cdot\pi}{4}\right)$$

Equation 10

or

$$\frac{BGB}{B0_i\cdot B0_f\cdot L} = R\left(\frac{-3\cdot\pi}{4}\right)\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots + \begin{bmatrix}0&-1\\1&0\end{bmatrix}\cdot\beta_f\right]\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots + \begin{bmatrix}1&0\\0&-1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots + \begin{bmatrix}0&1\\0&0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right)\ldots + \begin{bmatrix}0&0\\1&0\end{bmatrix}\cdot\left[\phi\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b}\right]\right]\cdot\left[\begin{bmatrix}1&0\\0&1\end{bmatrix}\ldots + \begin{bmatrix}0&-1\\1&0\end{bmatrix}\cdot\beta_i\right]\cdot R\left(\frac{3\cdot\pi}{4}\right)$$

Equation 11

which to zero and first order is approximated by

$$\begin{aligned}\frac{BGB}{B0_i \cdot B0_f \cdot L} = R\left(\frac{-3\cdot\pi}{4}\right)\cdot\Bigg[& \left[\left[\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix} + \begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\beta_f\right]\cdot\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}\cdot\left[\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix} + \begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\beta_i\right]\right] \ldots \\ & + \begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}\cdot\Bigg[\begin{bmatrix}1 & 0\\0 & -1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right) \ldots \\ & \quad + \begin{bmatrix}0 & 1\\0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\ & \quad + \begin{bmatrix}0 & 0\\1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2 - b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\Bigg]\cdot\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}\Bigg]\cdot R\left(\frac{3\cdot\pi}{4}\right)\end{aligned}$$

Equation 12

$$\begin{aligned}\frac{BGB}{B0_i \cdot B0_f \cdot L} = R\left(\frac{-3\cdot\pi}{4}\right)\cdot\Bigg[& \left[\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix} + \begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\left(\beta_f + \beta_i\right)\right] \ldots \\ & + \begin{bmatrix}1 & 0\\0 & -1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right) \ldots \\ & + \begin{bmatrix}0 & 1\\0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\ & + \begin{bmatrix}0 & 0\\1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2 - b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\Bigg]\cdot R\left(\frac{3\cdot\pi}{4}\right)\end{aligned}$$

Equation 13

$$\begin{aligned}\frac{BGB\cdot 2}{B0_i \cdot B0_f \cdot L} = \begin{bmatrix}-1 & 1\\-1 & -1\end{bmatrix}\cdot\Bigg[& \left[\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix} + \begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\left(\beta_f + \beta_i\right)\right] \ldots \\ & + \begin{bmatrix}1 & 0\\0 & -1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right) \ldots \\ & + \begin{bmatrix}0 & 1\\0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\ & + \begin{bmatrix}0 & 0\\1 & 0\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2 - b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\Bigg]\cdot\begin{bmatrix}-1 & -1\\1 & -1\end{bmatrix}\end{aligned}$$

Equation 14

or

$$\begin{aligned}\frac{BGB\cdot 2}{B0_i \cdot B0_f \cdot L} = \Bigg[& \left[\begin{bmatrix}-1 & 1\\-1 & -1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\1 & -1\end{bmatrix} + \begin{bmatrix}-1 & 1\\-1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & -1\\1 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\1 & -1\end{bmatrix}\cdot\left(\beta_f + \beta_i\right)\right] \ldots \\ & + \begin{bmatrix}-1 & 1\\-1 & -1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\0 & -1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\1 & -1\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right) \ldots \\ & + \begin{bmatrix}-1 & 1\\-1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\0 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\1 & -1\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\phi\cdot\cos(\chi)\right) \ldots \\ & + \begin{bmatrix}-1 & 1\\-1 & -1\end{bmatrix}\cdot\begin{bmatrix}0 & 0\\1 & 0\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\1 & -1\end{bmatrix}\cdot\left[\phi\cdot\frac{\left(a^2 - b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]\Bigg]\end{aligned}$$

Equation 15

$$\frac{BGB}{B0_i \cdot B0_f \cdot L} = \begin{bmatrix} \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot (\beta_f + \beta_i) \right] \dots \\ + \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \cdot \left(i \cdot \frac{a}{b} \cdot \phi \right) \dots \\ + \begin{bmatrix} -1 & 1 \\ -1 & 1 \end{bmatrix} \cdot \left(\frac{\sigma}{b} \cdot \frac{\phi}{2} \cdot \cos(\chi) \right) \dots \\ + \begin{bmatrix} -1 & -1 \\ 1 & 1 \end{bmatrix} \cdot \left[\frac{\phi}{2} \cdot \frac{(a^2 - b^2)}{\sigma \cdot \cos(\chi) \cdot b} \right] \end{bmatrix}$$

Equation 16

Remembering that

$$H = \begin{bmatrix} t_1 & s_2 \\ s_1 & t_2 \end{bmatrix} + \Delta\gamma \cdot t_1 \cdot \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$

Equation 17

$$F = \begin{bmatrix} t_2 & -s_1 \\ -s_2 & t_1 \end{bmatrix} + (\Delta\gamma + \Delta\theta) \cdot t_1 \cdot \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$

Equation 18

and letting $s_1 = s_2 = s$, $\zeta = t_2/t_1$ and $\Sigma = s/t_1$, the matrices, H and F may be written as

$$\frac{H}{t_1} = \begin{bmatrix} 1 & \Sigma \\ \Sigma & \zeta \end{bmatrix} + \Delta\gamma \cdot \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$

Equation 19

$$\frac{F}{t_1} = \begin{bmatrix} \zeta & -\Sigma \\ -\Sigma & 1 \end{bmatrix} + (\Delta\gamma + \Delta\theta) \cdot \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix}$$

Equation 20

The partial combined matrix, FBGBH, assuming identical polarisers, is then given by the equation

$$\frac{(F \cdot BGB \cdot H)}{L \cdot B0_i \cdot B0_f \cdot (t_1)^2} = \begin{bmatrix} \begin{bmatrix} \zeta & -\Sigma \\ -\Sigma & 1 \end{bmatrix} \dots \\ + (\Delta\gamma + \Delta\theta) \cdot \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix} \end{bmatrix} \cdot \begin{bmatrix} \left[\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \cdot (\beta_f + \beta_i) \right] \dots \\ + \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \cdot \left(i \cdot \frac{a}{b} \cdot \phi \right) \dots \\ + \begin{bmatrix} -1 & 1 \\ -1 & 1 \end{bmatrix} \cdot \left(\frac{\sigma}{b} \cdot \frac{\phi}{2} \cdot \cos(\chi) \right) \dots \\ + \begin{bmatrix} -1 & -1 \\ 1 & 1 \end{bmatrix} \cdot \left[\frac{\phi}{2} \cdot \frac{(a^2 - b^2)}{\sigma \cdot \cos(\chi) \cdot b} \right] \end{bmatrix} \cdot \begin{bmatrix} \begin{bmatrix} 1 & \Sigma \\ \Sigma & \zeta \end{bmatrix} \dots \\ + \Delta\gamma \cdot \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \end{bmatrix}$$

Equation 21

or, to first order

$$
\begin{aligned}
\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot BO_i\cdot BO_f\cdot\left(t_1\right)^2} = & \left[\begin{bmatrix}\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix}\ldots\\ +(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\right]\cdot\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix}+\Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\ldots\\
& +\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left(\beta_f+\beta_i\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & 1\\ -1 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\frac{\phi}{2}\cdot\cos(\chi)\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}-1 & -1\\ 1 & 1\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\cdot\left[\frac{\phi}{2}\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]
\end{aligned}
$$

Equation 22

$$
\begin{aligned}
\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot BO_i\cdot BO_f\cdot\left(t_1\right)^2} = & \left[\begin{bmatrix}\zeta & -\Sigma\\ -\Sigma & 1\end{bmatrix}+(\Delta\gamma+\Delta\theta)\cdot\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}\right]\cdot\left[\begin{bmatrix}1 & \Sigma\\ \Sigma & \zeta\end{bmatrix}+\Delta\gamma\cdot\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right]\ldots\\
& +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left(\beta_f+\beta_i\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\frac{\phi}{2}\cdot\cos(\chi)\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\frac{\phi}{2}\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]
\end{aligned}
$$

Equation 23

$$
\begin{aligned}
\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot BO_i\cdot BO_f\cdot\left(t_1\right)^2} = & \begin{bmatrix}\zeta+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot(\Sigma+\Delta\gamma) & \zeta\cdot(\Sigma+\Delta\gamma)+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot\zeta\\ -\Delta\theta & \zeta+(-\Sigma-\Delta\gamma-\Delta\theta)\cdot(\Sigma+\Delta\gamma)\end{bmatrix}\ldots\\
& +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left(\beta_f+\beta_i\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\frac{\phi}{2}\cdot\cos(\chi)\right)\ldots\\
& +\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\frac{\phi}{2}\cdot\frac{\left(a^2-b^2\right)}{\sigma\cdot\cos(\chi)\cdot b}\right]
\end{aligned}
$$

Equation 24

which is finally approximated to first order by

$$\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot BO_i\cdot BO_f\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta & \zeta\end{bmatrix}\ldots+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot(\beta_f+\beta_i)\ldots+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left(i\cdot\frac{a}{b}\cdot\phi\right)\ldots+\begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix}\cdot\left(\frac{\sigma}{b}\cdot\frac{\phi}{2}\cdot\cos(\chi)\right)\ldots+\begin{bmatrix}0 & 0\\ 1 & 0\end{bmatrix}\cdot\left[\frac{\phi}{2}\cdot\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b}\right]$$ Equation 25

$$\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot BO_i\cdot BO_f\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta+(\beta_f+\beta_i)+\frac{\phi}{2}\cdot\left[\frac{(a^2-b^2)}{\sigma\cdot\cos(\chi)\cdot b}-\frac{\sigma}{b}\cdot\cos(\chi)\right]+i\cdot\left(\frac{a}{b}\cdot\phi\right) & \zeta\end{bmatrix}$$

Equation 26

If the intersection angle of pump and probe beams is small, the terms a and b may be approximated by

$a=0$ Equation 27

and

$b=\sigma\cdot\cos(\chi)$ Equation 28

and the equation above simplifies to[1]

$$\frac{(F\cdot BGB\cdot H)}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot BO_i\cdot BO_f\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta+(\beta_f+\beta_i)-\phi & \zeta\end{bmatrix}$$ Equation 29

<u>which is an extension of the no birefringent element case</u>

$$\frac{(F\cdot G\cdot H)_{no\_birefringent\_element}}{e^{-\frac{\alpha_{av}}{2}\cdot\Lambda}\cdot e^{-i\cdot k_{av}\cdot\Lambda}\cdot(t_1)^2}=\begin{bmatrix}\zeta & 0\\ -\Delta\theta-\phi & \zeta\end{bmatrix}$$ Equation 30

[1] Note that, for both this case and in the equivalent case for an induced linear dichroism, there is no angular dependence associated with the birefringence factor.