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PhD thesis

A comprehensive study on cylindrical symmetry in optical physics:
Full-quantitative characterization of cylindrically polarized optical pulses

Masato Suzuki

Division of Applied Physics
Graduate School of Engineering, Hokkaido University

February 2016
# Contents

1 Introduction
   1.1 Cylindrical symmetry in optical physics .................. 1
   1.2 Methods for evaluating cylindrically polarized modes ...... 3
   1.3 Purpose of this research .................................. 4

2 Cylindrically polarized pulses ............................... 5
   2.1 Cylindrically polarized Laguerre-Gaussian modes ........... 5
      2.1.1 Derivation of cylindrically polarized Laguerre-Gaussian
            modes .................................................. 5
      2.1.2 Circularly polarized optical vortex basis ............... 7
      2.1.3 Polarization singularity ............................. 8
   2.2 Cylindrically polarized Laguerre-Gaussian pulses .......... 10
   2.3 Definition of terms describing light wave with polarization
       singularity .............................................. 11

3 Extended Stokes parameters and Pancharatnam-Berry phase 13
   3.1 Conventional Stokes parameters ............................ 13
      3.1.1 Definition .......................................... 13
      3.1.2 Degree of polarization .................................. 14
         A trivial approach .................................. 14
         Another approach from statistical optics or quantum
         mechanics .............................................. 15
      3.1.3 Poincaré sphere ....................................... 16
   3.2 Extended Stokes parameters for cylindrically polarized modes
       ......................................................... 17
   3.3 Degree of polarization of extended Stokes parameters ...... 19
      3.3.1 Definition ............................................. 19
      3.3.2 Degree of polarization defined for the spatial distribution 19
   3.4 Extended Poincaré sphere .................................. 20
   3.5 Comparison with other Stokes parameters .................... 21
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.6</td>
<td>Pancharatnam-Berry phase on an extended Poincaré sphere</td>
<td>23</td>
</tr>
<tr>
<td>3.6.1</td>
<td>“Hamiltonian” and Jones Matrix for motion on the surface of an extended Poincaré sphere</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>Obtaining a “Hamiltonian”</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>Jones matrix corresponding to $H'$</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>Hamiltonian for $q$-retarders</td>
<td>27</td>
</tr>
<tr>
<td>3.6.2</td>
<td>Berry connection and Berry phase on an extended Poincaré sphere</td>
<td>28</td>
</tr>
<tr>
<td>4</td>
<td>Generation and characterization of cylindrically polarized broadband pulses</td>
<td>31</td>
</tr>
<tr>
<td>4.1</td>
<td>Background</td>
<td>31</td>
</tr>
<tr>
<td>4.2</td>
<td>Concept of generating arbitrarily cylindrically polarized broadband pulse states</td>
<td>32</td>
</tr>
<tr>
<td>4.3</td>
<td>Experimental</td>
<td>33</td>
</tr>
<tr>
<td>4.3.1</td>
<td>Setup</td>
<td>33</td>
</tr>
<tr>
<td></td>
<td>Generation of broadband optical vortex pulses</td>
<td>34</td>
</tr>
<tr>
<td></td>
<td>Coherent combining system</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>Cylindrically polarized mode conversion</td>
<td>36</td>
</tr>
<tr>
<td></td>
<td>Finding the zero delay by using the spectral interference technique</td>
<td>37</td>
</tr>
<tr>
<td></td>
<td>Measuring polarization distributions</td>
<td>37</td>
</tr>
<tr>
<td>4.3.2</td>
<td>Results</td>
<td>38</td>
</tr>
<tr>
<td>4.4</td>
<td>Simulation</td>
<td>41</td>
</tr>
<tr>
<td>4.5</td>
<td>Discussion</td>
<td>43</td>
</tr>
<tr>
<td>5</td>
<td>Nonlinear propagation of axisymmetrically polarized pulses in an axisymmetrical system</td>
<td>47</td>
</tr>
<tr>
<td>5.1</td>
<td>Introduction of propagation of light wave in a uniaxial crystal</td>
<td>47</td>
</tr>
<tr>
<td>5.2</td>
<td>Linear propagation of axisymmetrically polarized beam in a uniaxial crystal</td>
<td>49</td>
</tr>
<tr>
<td>5.2.1</td>
<td>Paraxial linear wave equation</td>
<td>49</td>
</tr>
<tr>
<td>5.2.2</td>
<td>Solution to paraxial linear wave equation</td>
<td>50</td>
</tr>
<tr>
<td>5.3</td>
<td>Nonlinear propagation of axisymmetrically polarized pulses in a uniaxial crystal</td>
<td>53</td>
</tr>
<tr>
<td>5.3.1</td>
<td>Third-order nonlinear polarization</td>
<td>53</td>
</tr>
<tr>
<td>5.3.2</td>
<td>Paraxial nonlinear wave equation</td>
<td>54</td>
</tr>
<tr>
<td>5.4</td>
<td>Experimental</td>
<td>54</td>
</tr>
<tr>
<td>5.4.1</td>
<td>Setup</td>
<td>54</td>
</tr>
<tr>
<td>5.4.2</td>
<td>Results</td>
<td>56</td>
</tr>
<tr>
<td>5.5</td>
<td>Simulation</td>
<td>60</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

1.1 Cylindrical symmetry in optical physics

Symmetry plays a principal role in both of classical and modern physics [1, 2]. In optical physics, crystallite symmetry, which is microscopic structural symmetry, is a key to understand selection rules [3] and the forms of nonlinear susceptibility tensors [4] of nonlinear optical processes. Recently, the cylindrical symmetry of global polarization distribution of laser beams is an important topic for optical physics.

Linear polarized Gaussian beams (Fig. 1.1.1(a)), which is one of the conventional familiar laser beams, has $C_2$ symmetry of its polarization distribution whereas symmetry of its intensity distribution is $C_\infty$. In contrast to that, cylindrically polarized (CP) mode [5] is a laser beam mode whose polarization distribution has a rotational symmetry with respect to its beam axis.

Although a radially polarized (RP) mode, one of the typical CP modes having $C_\infty$ symmetry, was still generated in 1972 by Mushiake et al. [6], the CP modes came to attract attention gradually after the report on a improvement of laser cutting efficiency with the RP modes (Fig. 1.1.1(b)) by Niziev and Nesterov in 1999 [7] and calculations of tightly focused spot of axisymmetrically polarized (AxP) modes by Quabis et al. [8], and Youngworth and Brown [9] in 2000. There are respectively longitudinal electric or magnetic components whose spot size is beyond the diffraction limit when the RP or azimuthally polarized (AP) modes are tightly focused [9] (Fig. 1.1.2), which was experimentally confirmed by Dorn et al. [10]. Thereby, the RP and AP modes are expected to be applied to super-resolution microscopy [11, 12], particle acceleration [13, 14], laser processing [15–24], laser trapping [25–28], detection of single molecules [11] or dielectric nanoparticles [29], and
determination of orientation of nanoparticles \[30–32\] or point defects \[33\]. Furthermore, in 2015, it was found that the orientation of polarization distribution on the tightly focal spot of the superposition of CP modes forms Möbius strips \[34\]. Research on tightly focused CP modes will be accelerated.

Another important aspect of CP modes is the mode orthogonality \[5\]. That can be applied in mode-division multiplexing in optical communications \[35, 36\]. By propagating in free space or using a vortex fiber enabling CP modes to propagate \[37, 38\], some telecommunication experiments have been conducted \[35, 39\]. CP modes are also promising candidates for quantum communications and quantum information processing \[40–45\]. Taking notice of the relationship between CP Laguerre-Gaussian (LG) modes and
LG optical vortex (OV) [46] modes, some researchers utilize CP modes intermediately for generating OV modes [47, 48].

Some CP modes (for example, RP modes) contain all direction of linear polarization, which is recently utilized as high-speed kinematic sensing [49] and snapshot polarimetry [50–52]. AxP modes which have $C_\infty$ symmetry in polarization distributions match excitation of electron states in ring shaped materials such as mesoscopic rings [53] and ring shaped MX$_3$ quasi one dimensional crystals [54]. In addition to that, AxP beam is utilized for the optical tractor beam [55].

Hence, the CP modes having polarization singularity on the beam axes are about to involving many fields of physics and engineering as well as optics, and taking on increasing importance.

1.2 Methods for evaluating cylindrically polarized modes

Since the significance of CP modes is growing, numerous studies on generating CP beams and pulses have been conducted [56]. However, almost all studies yet adopt primitive and (partly-)qualitative methods in order to evaluate them:

- Comparing an image which is the experimentally captured intensity profile of a CP beam after a polarizer with an image which is a numerically calculated intensity profile [17, 57–85].

- Showing polarization distribution or spatial profiles of Stokes parameters experimentally obtained with the Stokes polarimetry method [18, 86–91].

Contamination of unwanted CP modes may result in a change of intensity profile at the tightly focused spot, cross talk in telecommunications, and accuracy deterioration of sensing and polarimetry. A full-quantitative evaluation for CP modes is important to sophisticate application mentioned above. Furthermore, it enables us to make a comparison among methods generating CP modes.

Higher-order Stokes parameters (HOSPs), which are independently invented by Millione et al. [92, 93] and Hollezek et al. [94]$^1$ at around the same time, are Stokes parameters for describing CP modes. The HOSPs do

$^1$Although Hollezek et al. call the parameters the hybrid Stokes parameters (HSPs) [94], we take HOSPs in this thesis.
not, however, comply with the framework of Stokes parameters, being calculated from a mode decomposition approach rather than from a statistical approach. Consequently, they are not capable for defining degree of polarization (DOP), which makes hard to make full-quantitative evaluations of generated CP beams and pulses experimentally.

The other approach to full-quantitatively evaluate CP modes is mode decomposition through the field reconstruction method [95]. Some research for evaluating OV pulses [96] and CP beams [97] have been conducted, but negative concomitants of the experimental set up is its complexity and instability because of using an interferometer.

Thus, a full-quantitatively evaluation method for experimentally generated CP modes with simple experimental set up is needed.

### 1.3 Purpose of this research

In order to establish a framework to measure CP modes full-quantitatively, we will introduce new extended Stokes parameters (ESPs) and a DOP complying with the framework of the conventional Stokes parameters (CSPs) and the DOP. We will show significance of the ESPs and their DOP through analyzing broadband CP pulses generated from a coherent combining system and CP pulses propagating in a nonlinear crystal.
Chapter 2

Cylindrically polarized pulses

2.1 Cylindrically polarized Laguerre-Gaussian modes

In this section, we introduce the cylindrically polarized Laguerre-Gaussian (CPLG) modes, following which we discuss the relationship between CPLG modes and circularly polarized LG OV modes, and their polarization singularity. We show an expression for CPLG pulses. Finally, the definition of words in this thesis is given.

2.1.1 Derivation of cylindrically polarized Laguerre-Gaussian modes

In this subsection, we obtain CPLG modes from the paraxial Helmholtz equation. In general, the electric field $E(r,t)$ ($r = (x, y, z)$) is a position vector and $t$ represents time) in vacuum obeys the wave equation [98]:

$$\nabla^2 E = \frac{1}{c^2} \partial_t E,$$

(2.1.1)

where $\nabla^2$ is the Laplacian and $c$ is the speed of light in vacuum. We focus on a (quasi-)monochromatic laser beam propagating in $+z$ direction, so we here use the paraxial approximation $|2ik\partial_z \tilde{E}| \gg |\partial_z^2 \tilde{E}|$, where $k$ is wavenumber and $\tilde{E}(r)$ is an envelope of the electric field oscillating at angular frequency $\omega(=ck)$: $E = \tilde{E} \exp\{i(kz - \omega t)\}$. We therefore acquire the paraxial Helmholtz equation

$$(\nabla_z^2 + 2ik\partial_z)\tilde{E} = 0,$$

(2.1.2)
where $\nabla^2_\perp$ is the transverse part of the Laplacian expressed by $\nabla^2_\perp = \partial^2_r + r^{-1}\partial_r + r^{-2}\partial^2_\phi$ in the cylindrical coordinates. We express the cylindrical coordinates as

$$r = re_r + \phi e_\phi + ze_z$$

(2.1.3)

$$\equiv \sqrt{x^2 + y^2} e_r + \arctan(y, x) e_\phi + ze_z,$$  

(2.1.4)

where $e_i$ ($i = r, \phi, z$) is the basis for cylindrical coordinate and $\arctan(y, x)$ is four-quadrant inverse tangent taking account of the signs of $y$ and $x$. Since the paraxial approximation means that divergence of the beam is enough small to neglect the $z$ component of the envelope of the electric field $\tilde{E}_z$. Thus, we consider only the envelope of the transverse electric field $\tilde{E}_\perp \equiv (\tilde{E}_x, \tilde{E}_y)^T$.

We here expressed the envelope of the electric field with the $l$th CP basis [99]:

$$e^l_r \equiv \begin{pmatrix} \cos(l\phi) \\ \sin(l\phi) \end{pmatrix}, \quad e^l_\phi \equiv \begin{pmatrix} -\sin(l\phi) \\ \cos(l\phi) \end{pmatrix}. \quad (2.1.5)$$

By using the linear combination of CP basis, the envelope of the transverse electric field is written by $\tilde{E}_\perp = \sum_l (u^l_r(r)e^l_r + u^l_\phi(r)e^l_\phi)$, the paraxial Helmholtz equation is expressed as

$$\left( e^l_r \mp ie^l_\phi \right) \left( \partial^2_r + \frac{1}{r}\partial_r - \frac{|l|^2}{r^2} + 2ik\partial_z \right) (u^l_r \pm iu^l_\phi) = 0. \quad (2.1.6)$$

Here, we utilize the method of separation of variables and assume that a solution can be written in the equation:

$$u^l_r \pm iu^l_\phi = L(\xi(z)) r^{|l|} \exp \left\{ i \left( Q(z) \frac{r^2}{2} + P(z) \right) \right\}. \quad (2.1.7)$$

We thus obtain the four following differential equations,

$$\left( \xi \partial^2_\xi + (|l| + 1 - \xi) \partial_\xi + p \right) L(\xi) = 0, \quad (2.1.8)$$

$$\partial_\xi Q(z) = -\frac{2\Re(Q(z))\xi}{k}, \quad (2.1.9)$$

$$\partial_\xi P(z) = \frac{i}{k} \{ Q(z)(|l| + 1) - 2p\Im(Q(z)) \}, \quad (2.1.10)$$

(2.1.11)

where $p$ is integer greater than or equal to 0, and $\Re(c)$ and $\Im(c)$ respectively stands for taking the real and the imaginary parts of $c$. Since the Eq. (2.1.8) is for the generalized Laguerre polynomials, $L(\xi) = L^{|l|}_p(\xi)$. The other functions
are calculated to be $\xi(z) = 2r^2/3(Q(z))$, $Q(z) = k/(z - i z_0)$, $P(z) = (1 + i z/z_0)^{-|l|-1}\exp(-(2p + |l| + 1)\arctan(z/z_0))$, where $z_0$ is called a Rayleigh length. Finally, a general solution to the Eq. (2.1.6) [5] is expressed by

$$\vec{E}_\perp(r, \phi, z) = \sum_{l,p} u_{l,p}^{\mathrm{CPLG}}(r, \phi, z) \left( \vec{u}_r^{l,p} e_r^l + \vec{u}_\phi^{l,p} e_\phi^l \right),$$

(2.1.12)

$$u_{l,p}^{\mathrm{CPLG}}(r, \phi, z) = \sqrt{\frac{2p!}{\pi (p + |l|)!}} \left( \frac{\sqrt{2}r}{w} \right)^{|l|} L_p^{|l|} \left( \frac{2r^2}{w^2} \right) \frac{w_0}{w} \times \exp \left\{ -\frac{r^2}{w^2} + i \left( \frac{k r^2}{2R} - \psi_{\mathrm{Gouy}}(z) \right) \right\},$$

(2.1.13)

where $u_{l,p}^{\mathrm{CPLG}}(r, \phi, z)$ is a CPLG mode indicated by the azimuthal index $l$ and the radial index $p$, and $\vec{u}_r^{l,p}, w_0, w = w(z)$ and $R = R(z)$ are respectively the amplitude for CPLG modes, the beam waist, a beam size at $z$ and a radius of curvature at $z$.

The phase factor $\psi_{\mathrm{Gouy}}(z)$ is named as the Gouy phase, which depends on the indices of $l$ and $p$:

$$\psi_{\mathrm{Gouy}}(z) = (|l| + 2p + 1)\arctan \left( \frac{z}{z_0} \right).$$

(2.1.17)

### 2.1.2 Circularly polarized optical vortex basis

In order to discuss the relationship between the CPLG modes and the circularly polarized LG OV modes, we introduce the circularly polarized OV basis:

$$e_+^{l_+} = \frac{e^{-il_+\phi}}{\sqrt{2}} \begin{pmatrix} 1 \\ +i \end{pmatrix}, \quad e_-^{l_-} = \frac{e^{il_-\phi}}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix},$$

(2.1.18)

where $l_+$ and $l_-$ are respectively topological charge of the left-circularly polarized (LCP) OV base $e_+^{l_+}$ and the right-circularly polarized (RCP) OV base $e_-^{l_-}$. When $l_+ = l_- = 0$, the LCP and the RCP OV basis are the conventional LCP base $e_+ = (1, +i)^T$ and the RCP base $e_- = (1, -i)^T$, respectively.
Here, we have the following simple relationship between the CP basis and the cylindrically polarized OV basis:

\[ e_{l \pm}^{l} = \frac{e_{l}^{l} \pm ie_{\phi}^{l}}{\sqrt{2}} \]  \hspace{1cm} (2.1.19)

This means that the envelope of the transverse electric field can be described as the linear combination of the cylindrically polarized OV basis.

\[ \tilde{E}_{\perp}(r, \phi, z) = \sum_{l, p} u_{CPLG}^{l,p} \left( \tilde{u}_{r}^{l,p} e_{l}^{l} + \tilde{u}_{\phi}^{l,p} e_{l}^{l} \right) = \sum_{l, p} \left( \tilde{u}_{r}^{l,p} u_{LG}^{l,p} e_{+}^{l} + \tilde{u}_{\phi}^{l,p} u_{LG}^{l,p} e_{-}^{l} \right), \]  \hspace{1cm} (2.1.20)

\[ \tilde{u}_{l \pm}^{l,p} = \frac{\tilde{u}_{r}^{l,p} \pm i\tilde{u}_{\phi}^{l,p}}{\sqrt{2}}, \]  \hspace{1cm} (2.1.21)

\[ u_{LG}^{l,p}(r, \phi, z) = u_{CPLG}^{l,p}(r, \phi, z)e^{\pm l\phi}, \]  \hspace{1cm} (2.1.22)

where \( u_{LG}^{l,p}(r, \phi, z) \) corresponds to a LG mode [46] typically called “an optical vortex”. Thus, the amplitude and phase distribution of a CPLG mode are same as those of a LG mode without the phase ramp around its beam axis.

### 2.1.3 Polarization singularity

We here discuss the polarization distribution and singularity of CPLG modes. Figure 2.1.1 shows typical polarization and intensity distributions of single CPLG modes. The intensity patterns of them are doughnut-shaped because there are singular points on the beam centers. In the field of optical physics, polarization patterns of light wave on the transverse planes can generally have polarization singularities [100–102], which are classified into V-points, C-points, L-points, and Σ-points [103]. The singular points of single CPLG modes can be classified into three categories; one is a V-point, another is a C-point and the other one is a phase singular point.

The V-point is the polarization singular point with respect to linear polarization, that is, linear polarized (LP) CPLG modes such as RP or AP LG modes. The C-point is also the polarization singular point for elliptical polarization as illustrated in Fig. 2.1.1(d). Vorticity of these singular points are characterized by the Stokes index \( \sigma_{12} \) [103]. We consider the complex Stokes field using the CSPs \( s_{1} \) and \( s_{2} \) (We will define them in Sec. 3.1.1.):

\[ \Sigma_{12}(r) = s_{1}(r) + is_{2}(r) = \sqrt{s_{1}(r)^{2} + s_{2}(r)^{2}} \exp(i\Phi_{12}), \]  \hspace{1cm} (2.1.23)
Figure 2.1.1: Intensity and polarization distributions of (a) a \((l, p) = (1, 0)\) RP LG mode, (b) a \((l, p) = (1, 1)\) RP LG mode, (c) \((l, p) = (1, 2)\) RP LG mode, (d) \((l, p) = (1, 0)\) CPLG mode, (e) \((l, p) = (1, 0)\) LCP LG mode, and (f) \((l, p) = (1, 0)\) RCP LG mode. V-points exist at the center of the RP LG modes in (a), (b) and (c). A C-point is located at the center of the CPLG mode in (d). Phase singularly points are on the center of the LCP and the RCP modes in (e) and (f), respectively.

where \(\Phi_{12}\) is the angle of orientation of linear polarization or the long axis of elliptic polarization from +x axis. The Stokes index is defined by

\[
\sigma_{12} = \frac{1}{2\pi} \oint_C \nabla \Phi_{12} \cdot ds = \begin{cases} 
  l & (C \text{ contains the beam axis}), \\
  0 & (\text{otherwise})
\end{cases}
\]  

(2.1.24)

where \(ds\) is a linear element on a closed loop \(C\).

The other singularity, the phase singularity appears with regard to circularly polarized LG modes. Since the circularly polarized state is axisymmetric, its polarization states at the center can be determined. However, the value of the phase at the center is undetermined, which gives the phase singular point. The vorticity of it is the topological charge \(l\). Thus, it is also interpreted that a circularly polarized LG mode is circular polarized OV with topological charge \(l\).
A single CPLG mode therefore has a singular point with vorticity \( l \) on its beam axes. The radial index \( p \) of a single CPLG mode is connected to the number of node on the intensity distribution as that of a single LG OV mode is \([104]\).

### 2.2 Cylindrically polarized Laguerre-Gaussian pulses

Optical pulses are basically interpreted as the electric field whose envelope is time-dependent. In general, the transverse electric field \( E_\perp(r,t) \) is expressed by the Fourier transform of a product of a complex spectrum amplitude \( \tilde{E}_\perp^{(\omega)}(r,\omega) \) and a term of oscillation \( \exp\{i(kz - \omega t)\} \) with respect to the angular frequency \([105]\):

\[
E_\perp(r,t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{E}_\perp^{(\omega)}(r,\omega) \exp(-i\omega t) d\omega. \tag{2.2.1}
\]

Since \( \tilde{E}_\perp^{(\omega)}(r,\omega) \exp(-ikz) \) satisfies Eq. (2.1.2), it can be expressed as the superposition of CPLG modes.

\[
\tilde{E}_\perp^{(\omega)}(r,\omega) = \sum_{l,p} u_{l,p}^{\text{CPLG}}(r,\phi, z, k) \left( \tilde{u}_r^{l,p}(k)e_r^l + \tilde{u}_\phi^{l,p}(k)e_\phi^l \right) \exp(ikz), \tag{2.2.2}
\]

where the angular frequency \( \omega \) and the wavenumber \( k \) are connected by the dispersion relationship (in vacuum, \( k(\omega) = \omega/c \)). The CPLG modes \( u_{l,p}^{\text{CPLG}} \) depend on the angular frequency, however, we can neglect the dependence when a Ti:Sapphire (Ti:Sa) laser amplifier (spectrum range is from \( \sim 780 \) to \( \sim 820 \) nm) is used in our experiments. The angular frequency dependence of the beam waist \( w_0 = \sqrt{2z_0/k(\omega)} = \sqrt{z_0\lambda/\pi} \) causes that of the CPLG modes. Figure 2.2.1 legitimatizes the neglect since the beam waist changes by a few percent in the spectral range of the Ti:Sa laser amplifier.

Thus, unless otherwise specified we hereafter use the expression of

\[
\tilde{E}_\perp^{(\omega)}(r,\omega) = f_\omega(\omega) \exp(i(k(\omega)z) \sum_{l,p} u_{l,p}^{\text{CPLG}}(r,\phi, z) \left( \tilde{u}_r^{l,p}e_r^l + \tilde{u}_\phi^{l,p}e_\phi^l \right), \tag{2.2.3}
\]

\[
E_\perp(r,t) = f(t,z) \tilde{E}_\perp(r)
= f(t,z) \sum_{l,p} u_{l,p}^{\text{CPLG}}(r,\phi, z) \left( \tilde{u}_r^{l,p}e_r^l + \tilde{u}_\phi^{l,p}e_\phi^l \right), \tag{2.2.4}
\]

where \( f(t,z) = (2\pi)^{-1} \int_{-\infty}^{\infty} f_\omega(\omega) \exp\{i(k(\omega)z - \omega t)\} d\omega. \)
Figure 2.2.1: The wavelength dependence of the beam waist $w_0/\sqrt{z_0} = \sqrt{\lambda/\pi}$.

2.3 Definition of terms describing light wave with polarization singularity

We define the terms describing light wave with polarization or phase singularity in this thesis since they have not been rigidly used among many scientists. We consider the condition given by the following equation in order to describe the terms with easy mathematical expression:

$$\tilde{E}(r, \phi, z) \propto \{A_+ e^{l_+} + A_- e^{l_-}\} e^{il^'\phi},$$

(2.3.1)

where, $A_\pm$ are amplitudes. We omit an insignificance case $A_+ = A_- = 0$.

**Light wave with polarization singularity**

Light wave having $V$-points or $C$-points.

**Light wave with phase singularity**

Light wave with phase dislocations.

**Vector vortex**

$l \neq 0$. (sometimes referred to as “cylindrical vector modes”)

**CPLG modes**

$l \neq 0$ and $l' = 0$.

**AxP modes**

$l = 1$ and $l' = 0$.

**LP CPLG modes**

$l \neq 0$, $l' = 0$ and $|A_+|^2 - |A_-|^2 = 0$. 

11
Figure 2.3.1: Definition of terms in this thesis.

<table>
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<tr>
<th>Mode Type</th>
<th>Condition</th>
<th>Characteristics</th>
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<tr>
<td>RP LG modes</td>
<td>( l = 1, l' = 0 ) and ( A_+ = A_- ).</td>
<td></td>
</tr>
<tr>
<td>AP LG modes</td>
<td>( l = 1, l' = 0 ) and ( A_+ = -A_- ).</td>
<td></td>
</tr>
<tr>
<td>Circularly polarized LG (OV) modes</td>
<td>( l \neq 0, l' = 0 ), and ( A_+ = 0 ) or ( A_- = 0 ).</td>
<td></td>
</tr>
<tr>
<td>LG OV modes</td>
<td>( l = 0 ) and ( l' \neq 0 ).</td>
<td></td>
</tr>
<tr>
<td>Uniformly polarized LG modes</td>
<td>( l = 0 ).</td>
<td></td>
</tr>
</tbody>
</table>

Figure 2.3.1 gives graphic explanation of the terms. In this thesis, we focus on the CPLG modes.
Chapter 3

Extended Stokes parameters
and Pancharatnam-Berry phase

In this chapter, we introduce the ESPs which is capable for definition of DOP unlike HOSPs. First, we will make a brief review of the CSPs, the conventional DOP and the conventional Poincaré sphere (CPS) in order to compare with the ESPs, their DOP and the extended Poincaré sphere (EPS), respectively. After that, we define the ESPs, their DOP and the EPS. We also make a comparison with the other Stokes parameters which have been proposed. We discuss the Pancharatnam-Berry phase (PBP) on the EPS at the end of this chapter.

3.1 Conventional Stokes parameters

3.1.1 Definition

The CSPs are the measurable parameters [106] that describe the spatially homogeneous polarization state of a transverse electric field as follows:

\[ \mathbf{E}_\perp(r,t) = g(r) \mathbf{\bar{E}}_\perp(t) = g(r) \begin{pmatrix} \bar{E}_x(t) \\ \bar{E}_y(t) \end{pmatrix}. \] (3.1.1)

The CSPs are defined by the “expectation value” of the Pauli spin matrices with respect to the time axis [107]:

\[ \mathbf{S} = (\mathbf{E}_\perp^\dagger \mathbf{\sigma}^{xy} \mathbf{E}_\perp)_t. \] (3.1.2)

Here, the conventional Stokes vector and the Pauli spin matrix for \( x \) and \( y \) basis are written as

\[ \mathbf{S} = (S_0, S_1, S_2, S_3)^T. \quad \mathbf{\sigma}^{xy} = (\sigma_0, \sigma_3, \sigma_1, \sigma_2)^T, \] (3.1.3)
where \( \sigma_i (i = 0 - 3) \) are defined \[108\] by
\[
\sigma_0 \equiv \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \sigma_1 \equiv \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \sigma_2 \equiv \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \sigma_3 \equiv \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.
\]
(3.1.4)

The symbol \( \langle \cdots \rangle_t \) represents the time average;
\[
\langle f(t) \rangle_t = \lim_{T \to \infty} \frac{1}{T} \int_0^T f(t) dt.
\]
(3.1.5)

The CSPs can be applied to the spatially inhomogeneous polarization states. In this case, we divide the transverse electric field into mesh and assume the polarization state is uniform in each area. Thereby, we can acquire the polarization distribution of the inhomogeneous polarized states (see Sec. A.3) through the spatially dependent CSPs. We here introduce the CSPs at the position \( r \) as follows:
\[
s(r) = (s_0(r), s_1(r), s_2(r), s_3(r))^T = \langle \bar{E}_\perp(r) \sigma_{xy} \bar{E}_\perp(r) \rangle_t.
\]
(3.1.6)

### 3.1.2 Degree of polarization

In this section, we introduce the DOP, which tells us how the instantaneous polarization state is stationary. We introduce them in two of the ways.

**A trivial approach**

The first is a trivial approach \[109, 110\]. The CSPs have the following relationship:
\[
S_0 \geq |S_i| \quad (i = 1 - 3),
\]
(3.1.7)

which means that the sum of squares of the normalized CSPs \( \tilde{S}_i = \frac{S_i}{S_0} \) \( (i = 1 - 3) \) satisfies
\[
0 \leq \sqrt{\tilde{S}_1^2 + \tilde{S}_2^2 + \tilde{S}_3^2} \leq 1
\]
(3.1.8)

Here, we define the DOP as
\[
\mathcal{P} = \sqrt{\tilde{S}_1^2 + \tilde{S}_2^2 + \tilde{S}_3^2},
\]
(3.1.9)

thus, we obtain that \( 0 \leq \mathcal{P} \leq 1 \).

By using the DOP, we can split a Stokes vector into two vectors:
\[
S = \begin{pmatrix} S_0 \mathcal{P} \\ \frac{S_1}{S_0} \\ \frac{S_2}{S_0} \\ \frac{S_3}{S_0} \end{pmatrix} + \begin{pmatrix} S_0(1 - \mathcal{P}) \\ 0 \\ 0 \\ 0 \end{pmatrix},
\]
(3.1.10)
where the first and the second terms are respectively called the perfect polarized and the unpolarized Stokes vectors [109]. If $P = 1$ or 0, the Stokes vector respectively have only the perfect polarized part or the unpolarized part, which means that the polarization state is full stationary or unstable. When $0 < P < 1$, the Stokes vector has both of the perfect polarized and the unpolarized Stokes vectors. That state is called the partially polarized state, which is a polarization state of neither full stationary nor unsteadiness.

Another approach from statistical optics or quantum mechanics

Another approach is using the coherency matrix [111, 112] or the density matrix of two level systems [108] in the context of statistical optics or quantum mechanics, respectively. The coherency matrix is introduced [111] by

$$J = (\langle \bar{E} E^\dagger \rangle_t) = \begin{pmatrix} \langle |E_x|^2 \rangle_t & \langle E_x E_y^* \rangle_t \\ \langle E_y^* E_x \rangle_t & \langle |E_y|^2 \rangle_t \end{pmatrix} = \frac{1}{2} \begin{pmatrix} S_0 + S_1 & S_2 - iS_3 \\ S_2 + iS_3 & S_0 - S_1 \end{pmatrix} = S \cdot \sigma^{xy}. \quad (3.1.11)$$

Satisfying $J = J^\dagger$, the coherent matrix is an Hermite matrix. Thus, the matrix is diagonalizable by a unitary matrix $P$:

$$PJ^\dagger P^\dagger = \begin{pmatrix} \lambda_+ & 0 \\ 0 & 0 \end{pmatrix}, \quad (3.1.12)$$

where $\lambda_\pm$ are the eigenvalues written as

$$\lambda_\pm = \frac{\text{tr}[J]}{2} \left( 1 \pm \sqrt{1 - 4 \frac{\text{det}[J]}{\text{tr}[J]^2}} \right) = \frac{1}{2} S_0 \pm \sqrt{S_1^2 + S_2^2 + S_3^2} = \frac{S_0}{2} (1 \pm P). \quad (3.1.13)$$

Here, the DOP is defined [111] by

$$P = \sqrt{1 - 4 \frac{\text{det}[J]}{\text{tr}[J]^2}} (= \sqrt{S_1^2 + S_2^2 + S_3^2}). \quad (3.1.14)$$

By using the DOP $P$, the coherency matrix is described as

$$PJ^\dagger P = S_0 \begin{pmatrix} P & 0 \\ 0 & 0 \end{pmatrix} + \frac{S_0}{2} \begin{pmatrix} 1 - P & 0 \\ 0 & 1 - P \end{pmatrix}, \quad (3.1.15)$$

$$\text{tr}[PJ^\dagger P] = \text{tr}[J] = S_0 P + S_0 (1 - P). \quad (3.1.16)$$

The first and the second terms are the perfect polarized and the unpolarized states, respectively.
Since the coherency matrix is quite similar to the density matrix exclusive of the time average in quantum mechanics, we can make the analogy between them. Equation (3.1.16) is interpreted as the sum of population, thus the energy ratio between the perfect polarized and the unpolarized states is given by $P : 1 - P$, which is the same ratio in the trivial approach.

The partially polarized and the unpolarized states are considered to be the mixed states. For example, a mixed state of the $x$-polarized and $y$-polarized states is written as

$$J_{\text{mixed}} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \rho_{xx} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \rho_{yy} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \rho_{xx} \geq \rho_{yy} \\ \rho_{xx} < \rho_{yy} \end{pmatrix},$$

where $\rho_{xx}$ and $\rho_{yy}(= 1 - \rho_{xx})$ are respectively the population of $x$- and $y$-polarized states. When $\rho_{xx} = 1$ or $\rho_{yy} = 1$, the polarization state is a perfect polarized state. When $\rho_{xx} < 1$, which is a mixed state of $x$- and $y$-polarized states, the DOP is less than 1 and consequently the polarization state is a partially polarized state ($\rho_{xx} \neq \rho_{yy}$) or unpolarized state ($\rho_{xx} = \rho_{yy} = 0.5$).

### 3.1.3 Poincaré sphere

The CPS is a sphere for describing a polarization state. We introduce the normalized Stokes vector:

$$\tilde{S} = \begin{pmatrix} \tilde{S}_1 \\ \tilde{S}_2 \\ \tilde{S}_3 \end{pmatrix}.$$ (3.1.18)

Any homogeneous polarization states can be described by points (or vectors) in the space of $(\tilde{S}_1, \tilde{S}_2, \tilde{S}_3)$. The length of the normalized Stokes vector is equal to the DOP, thus the vector does not exceed the surface of the unit sphere. We call this sphere (conventional) Poincaré sphere (CPS) [110]. When the polarization state is the perfect polarized, the normalized Stokes vector touches the surface of the CPS. Otherwise, the normalized Stokes vector is in the CPS.

Figure 3.1.1 shows the CPS. Each perfect polarized state corresponds to each point on the surface of the CPS one-on-one. By introducing the
spherical coordinates \((r_{ps}, \theta_{ps}, \phi_{ps})\), the Stokes vector is written as
\[
\mathbf{S} = \mathcal{P} \begin{pmatrix} 
\sin \theta_{ps} \cos \phi_{ps} \\
\sin \theta_{ps} \sin \phi_{ps} \\
\cos \theta_{ps}
\end{pmatrix}
\] (3.1.19)

where \(r_{ps} = \mathcal{P}\).

### 3.2 Extended Stokes parameters for cylindrically polarized modes

In this section, we newly introduce the ESPs for CP modes. We decompose the transverse electric vector into the \(l\)th CP components \(E^l_r\) and \(E^l_\phi\) as follows:
\[
\mathbf{E}_\perp(r,t) = R_{l\phi} \mathbf{E}_\perp^l = R_{l\phi} \begin{pmatrix} 
E^l_r(r,t) \\
E^l_\phi(r,t)
\end{pmatrix} = E^l_r(r,t) e^l_r + E^l_\phi(r,t) e^l_\phi, \quad (3.2.1)
\]

where
\[
R_{\phi} = \begin{pmatrix} 
\cos \theta & -\sin \theta \\
\sin \theta & \cos \theta
\end{pmatrix}
\] (3.2.2)

represents the rotation matrix.
The ESPs are defined by

\[ S_{l}^{E} = \langle E_{\perp}^{\dagger} R_{\phi} \sigma^{xy} R_{-\phi} E_{\perp} \rangle_{\perp, t} \]  \hspace{2cm} (3.2.3)

\[ = \langle \bar{E}_{\perp}^{\dagger} \sigma^{xy} \bar{E}_{\perp} \rangle_{\perp, t}, \]  \hspace{2cm} (3.2.4)

where the extended Stokes vector (ESV) \( S_{l}^{E} \) is given by

\[ S_{l}^{E} = (S_{0,l}^{E}, S_{1,l}^{E}, S_{2,l}^{E}, S_{3,l}^{E})^{T} \quad (3.2.5) \]

and the symbol \( \langle \cdot \cdot \cdot \rangle_{\perp, t} \) means average in time and in the beam cross section \( A \):

\[ \langle f(r, t) \rangle_{\perp, t} = \lim_{T \to \infty} \frac{1}{AT} \int_{A} \int_{0}^{T} f(r, t) dt dx dy. \]  \hspace{2cm} (3.2.6)

Since Eq (3.2.4) complies with the definition of the CSPs (Eq. (3.1.2)), we can say that the ESPs are truly extensions of the CSPs from the temporal to the spatiotemporal region [99].

The ESPs relate to the CSPs at the position \( r \). By using \( \bar{E}_{\perp}^{l}(r, t) \), the CPS \( s(r) \) is described as follows:

\[ s(r) = (s_{0}(r), s_{1}(r), s_{2}(r), s_{3}(r))^{T} \]
\[ = \langle E_{\perp}^{\dagger} \sigma^{xy} E_{\perp} \rangle_{l} \]
\[ = \langle E_{\perp}^{\dagger} R_{-\phi} \sigma^{xy} R_{\phi} \bar{E}_{\perp} \rangle_{l} \]
\[ = \begin{pmatrix}
    \langle \bar{E}_{\perp}^{\dagger} \sigma_{0} \bar{E}_{\perp} \rangle_{l} \\
    \cos 2l\phi \langle \bar{E}_{\perp}^{\dagger} \sigma_{3} \bar{E}_{\perp} \rangle_{l} - \sin 2l\phi \langle \bar{E}_{\perp}^{\dagger} \sigma_{1} \bar{E}_{\perp} \rangle_{l} \\
    \sin 2l\phi \langle \bar{E}_{\perp}^{\dagger} \sigma_{3} \bar{E}_{\perp} \rangle_{l} + \cos 2l\phi \langle \bar{E}_{\perp}^{\dagger} \sigma_{1} \bar{E}_{\perp} \rangle_{l} \\
    \langle \bar{E}_{\perp}^{\dagger} \sigma_{2} \bar{E}_{\perp} \rangle_{l}
\end{pmatrix}
\]
\[ (i = 0 - 3). \]  \hspace{2cm} (3.2.7)

Spatially averaging the both sides of Eq. (3.2.7), we acquire the relationship between the ESPs and the CSPs:

\[ S_{l}^{E} = \begin{pmatrix}
    \langle s_{0}(r) \rangle_{\perp} \\
    \langle \cos 2l\phi \ s_{1}(r) + \sin 2l\phi \ s_{2}(r) \rangle_{\perp} \\
    \langle -\sin 2l\phi \ s_{1}(r) + \cos 2l\phi \ s_{2}(r) \rangle_{\perp} \\
    \langle s_{3}(r) \rangle_{\perp}
\end{pmatrix}, \]  \hspace{2cm} (3.2.8)
where the symbol $\langle \cdots \rangle_\perp$ represents the average in the beam cross section written by
\[
\langle f(r) \rangle_\perp = \frac{1}{A} \int_A \int_A f(r) \, dx \, dy.
\]

Equation (3.2.8) is an important relationship in order to experimentally obtain the values of the ESPs by using polarizing optical elements and a charge coupled device (CCD) camera. The experimental technique for obtaining $s(r)$ is described in Appendix A.

3.3 Degree of polarization of extended Stokes parameters

3.3.1 Definition

By extending Eq. (3.1.11) from the temporal average to the spatiotemporal average, we define the coherency matrix for $l$th CP components as
\[
J_l = \langle \vec{E}_l^r \phi \rangle_{\perp,t} = \left( \begin{array}{cc}
\langle |E_l^r|^2 \rangle_{\perp,t} & \langle E_l^r E_l^\phi \rangle_{\perp,t} \\
\langle E_l^r E_l^\phi \rangle_{\perp,t} & \langle |E_l^\phi|^2 \rangle_{\perp,t}
\end{array} \right) = \frac{1}{2} \left( S_{0,l}^E + S_{1,l}^E S_{2,l}^E - i S_{3,l}^E \right) \sigma_{xy},\]

which complies the definition of the conventional DOP (Eq. (3.1.14)).

We name $J_l$ the $l$th extended coherency matrix. By using $J_l$, we derive the $l$th DOP of the $l$th ESPs as [99]
\[
\mathcal{P}_l = \sqrt{1 - 4 \det[J_l] (\text{tr}[J_l])^2} = \sqrt{(S_{1,l}^E)^2 + (S_{2,l}^E)^2 + (S_{3,l}^E)^2},
\]

which complies the definition of the conventional DOP (Eq. (3.1.14)).

Since the conventional DOP is based on the time average, it represents the stability of polarization states with respect to the time axis. In contrast to that, the $l$th DOP comes from the spatiotemporal average, and thus it expresses the stability of $l$th CP states with respect to the time axis and the spatial axes in the beam cross section.

3.3.2 Degree of polarization defined for the spatial distribution

The $l$th DOP $\mathcal{P}_l$ is not useful in experimental research because it contains unpolarized part both along the time axis and the the spatial axes in the
beam cross section. In this thesis, we focus on the polarization distribution of the $l$th CP pulses, we, therefore, introduce another parameter expressing the symmetry of the polarization distribution in the beam cross section:

$$\mathcal{P}_{l}^{\text{space}} = \sqrt{(S_{E 1, l}^{E})^2 + (S_{E 2, l}^{E})^2 + (S_{E 3, l}^{E})^2} / S_{0, l}^{E, (P)}, \quad (3.3.3)$$

where $S_{0, l}^{E, (P)}$ is an amount proportional to the energy or power in the beam cross section of the perfect polarized part with respect to time:

$$S_{0, l}^{E, (P)} = \langle \sqrt{(s_1(r))^2 + (s_2(r))^2 + (s_3(r))^2} \rangle _{\perp}. \quad (3.3.4)$$

We call $\mathcal{P}_{l}^{\text{space}}$ the degree of polarization defined for the spatial distribution (DOP-SD) [99].

The ESV is divided into three vectors as follows:

$$S_{l}^{E} = \begin{pmatrix} S_{0, l}^{E} - S_{0, l}^{E, (P)} \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} S_{0, l}^{E, (P)} (1 - \mathcal{P}_{l}^{\text{space}}) \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} S_{0, l}^{E, (P)} \mathcal{P}_{l}^{\text{space}} \\ S_{1, l}^{E} \\ S_{2, l}^{E} \end{pmatrix}. \quad (3.3.5)$$

The first, second and third terms respectively represent temporally unpolarized (TU), temporally-perfect-polarized but spatially unpolarized (TPPSU), and temporally- and spatially-perfect-polarized (TSPP) ESVs [113].

### 3.4 Extended Poincaré sphere

As is the case of the CSPs, we can define the EPS for the ESPs. In contrast to the CSPs, there are three types of the EPSs depending on the ways of normalization of ESV.

The first EPS, “the usual EPS”, is defined by the normalized ESV by $S_{0, l}^{E}$:

$$\hat{S}_{l}^{E} = \begin{pmatrix} \hat{S}_{1, l}^{E} \\ \hat{S}_{2, l}^{E} \\ \hat{S}_{3, l}^{E} \end{pmatrix} \equiv \frac{1}{S_{0, l}^{E}} \begin{pmatrix} S_{1, l}^{E} \\ S_{2, l}^{E} \\ S_{3, l}^{E} \end{pmatrix}. \quad (3.4.1)$$

By using the spherical coordinate $(R_{l}^{E}, \theta_{l}^{E}, \phi_{l}^{E})$ of the $l$th EPS, $\hat{S}_{l}^{E}$ is written by

$$\hat{S}_{l}^{E} = \mathcal{P}_{l} \begin{pmatrix} \sin \theta_{l}^{E} \cos \phi_{l}^{E} \\ \sin \theta_{l}^{E} \sin \phi_{l}^{E} \\ \cos \theta_{l}^{E} \end{pmatrix}, \quad (3.4.2)$$
which means that the distance between the origin and \( S^E_1 \) is the \( l \)th DOP \( P_l \).

The second EPS, “the spatial EPS”, is written by the normalized ESV by \( S^E_0 \),
\[
\tilde{S}^E_{l(P)} = \frac{1}{S_{0,l}} \begin{pmatrix} S^E_{1,l} \\ S^E_{2,l} \\ S^E_{3,l} \end{pmatrix} = P^\text{space}_l \begin{pmatrix} \sin \theta^E_l \cos \phi^E_l \\ \sin \theta^E_l \sin \phi^E_l \\ \cos \theta^E_l \end{pmatrix} 
\]
(3.4.3)
The length of \( \tilde{S}^E_{l(P)} \) equals to \( P^\text{space}_l \).

The third EPS, “the TSPP EPS”, is described by the normalized TSPP ESV:
\[
\tilde{S}^E_{l,\text{TSPP}} = \frac{1}{S_{0,l} P^\text{space}_l} \begin{pmatrix} S^E_{1,\text{TSPP}} \\ S^E_{2,\text{TSPP}} \\ S^E_{3,\text{TSPP}} \end{pmatrix} = \begin{pmatrix} \sin \theta^E_l \cos \phi^E_l \\ \sin \theta^E_l \sin \phi^E_l \\ \cos \theta^E_l \end{pmatrix}, \quad (3.4.4)
\]
which corresponds to the normalized HOSPs [92–94]. The point described by \( \tilde{S}^E_{l,\text{TSPP}} \) is always on the \( l \)th TSPP EPS (Fig. 3.4.1).

### 3.5 Comparison with other Stokes parameters

As mentioned in Sec. 1.2, some researchers have proposed the concept, named the HOSPs [92–94], similar to the ESPs. In this section, we compare the ESPs with the HOSPs.
The framework of the HOSPs is described by one of extensions of the Stokes parameters. These parameters are essentially defined by two coefficients of two states. By assuming that the transverse electric field can be express as follows:

\[
E_{\perp}(r, t) = g(r, t) \left( c_+^l e_+^l + c_-^l e_-^l \right),
\]

the \( l \)th HOSPs \( S_l^{\text{HOSP}} \) and their coherency matrix \( J_l^{\text{HOSP}} \) are respectively written by \[92, 94\]

\[
S_l^{\text{HOSP}} = \begin{pmatrix} (c_+^l)^* & (c_-^l)^* \end{pmatrix} \begin{pmatrix} c_+^l \\ c_-^l \end{pmatrix},
\]

\[
J_l^{\text{HOSP}} = \begin{pmatrix} |c_+^l|^2 & c_+^l c_-^l^* \\ c_+^l c_-^l & |c_-^l|^2 \end{pmatrix} = S_l^{\text{HOSP}} \cdot \sigma,
\]

where

\[
\sigma = (\sigma_0, \sigma_1, \sigma_2, \sigma_3)^T.
\]

Since

\[
\sqrt{1 - 4 \det[J_l^{\text{HOSP}}]} / (\text{tr}[J_l^{\text{HOSP}}])^2 = 1
\]

always satisfies, we can say that the DOP for the HOSPs has hardly information describing the light states. In that context, the DOP for the HOSPs is undefinable (Table 3.5.1) while that the DOP for the ESPs is definable.

Table 3.5.1: Comparison among Stokes parameters

<table>
<thead>
<tr>
<th></th>
<th>CSPs</th>
<th>ESPs[99, 114]</th>
<th>HOSPs[92–94]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Essence</strong></td>
<td>a coherency matrix averaged in time</td>
<td>a coherency matrix averaged in time and space</td>
<td>a coherency matrix made by two mode coefficients</td>
</tr>
<tr>
<td><strong>DOP</strong></td>
<td>definable</td>
<td>definable</td>
<td>undefinable</td>
</tr>
<tr>
<td>( l )th CP states with ( P_l = 1 )</td>
<td>unrepresentable except ( l = 0 )</td>
<td>representable</td>
<td>representable</td>
</tr>
<tr>
<td>( l )th CP states with ( P_l \neq 1 ) and ( P_0^{\text{space}} = 1 )</td>
<td>unrepresentable except ( l = 0 )</td>
<td>representable</td>
<td>unrepresentable</td>
</tr>
</tbody>
</table>
3.6 Pancharatnam-Berry phase on an extended Poincaré sphere

The PBP is one of the geometrical phases [115] which exist in various physical systems of quantum mechanics [116, 117], classical mechanics [118] and optics [119–123].

We here discuss the PBP on an EPS. First, we acquire a general “Hamiltonian” and a corresponding Jones matrix expressing an optical element that make a trajectory on an EPS. Then, we apply the Berry connection [116] to the “Hamiltonian” in order to obtain the PBP on an EPS.

3.6.1 “Hamiltonian” and Jones Matrix for motion on the surface of an extended Poincaré sphere

Obtaining a “Hamiltonian”

Any Hermite operator $\hat{H}$ is written [108] as

$$\hat{H} = \frac{1}{2} \mathbf{H} \cdot \sigma^{xy}, \quad (3.6.1)$$

where every element of $\mathbf{H} = (H_0 \ H_1 \ H_2 \ H_3)^{T}$ represents a coefficient of Pauli matrices $\sigma^{xy}_i$. By analogy in quantum mechanics, the “Schrödinger equation” for $e^{-iH_0z/2} \bar{E}_\perp$ is expressed [108] by

$$i \frac{d}{dz} (e^{-iH_0z/2} \bar{E}_\perp) = \hat{H} e^{-iH_0z/2} \bar{E}_\perp, \quad (3.6.2)$$

where $e^{-iH_0z/2}$ corresponds to the dynamical phase term [116]. This equation is transformed into the equation without the dynamical phase term$^1$:

$$i \frac{d}{dz} \bar{E}_\perp = \hat{H}' \bar{E}_\perp, \quad (3.6.3)$$

where

$$\hat{H}' = \begin{pmatrix} H_1 & H_2 - iH_3 \\ H_2 + iH_3 & -H_1 \end{pmatrix} = \hat{H} \cdot \tilde{\sigma}^{xy} = \begin{pmatrix} \sigma_3 \\ \sigma_1 \end{pmatrix}, \quad (3.6.4)$$

$^1$This equation can be regarded as one of the Maxwell-Schrödinger equations [124, 125].
From Eq. (3.3.1) and Eq. (3.6.3), we acquire

\[
\frac{i}{d} \frac{d}{dz} J_l = i \left( \left\langle \frac{dE_l^\perp}{dz} \frac{dE_l^{\perp\dagger}}{dz} \right\rangle_{\perp,t} + i \left\langle \frac{E_l^\perp}{dz} \right\rangle_{\perp,t} \right)
= \langle \hat{H}' \frac{dE_l^\perp}{dz} \rangle_{\perp,t} - \langle \frac{dE_l^{\perp\dagger}}{dz} \hat{H}' \rangle_{\perp,t}
= [\hat{H}', J_l],
\]
(3.6.5)

which is the Liouville–von Neumann equation [126] for the “Hamiltonian” \( \hat{H}' \).

We consider the light beam with \( S_{0,l}^E = 1 \) and \( \mathcal{P}_l = \mathcal{P}_{l \text{space}} = 1 \), which means that the normalized ESV \( \tilde{S}_l^E \) touch on the surface of the \( l \)th EPS. The \( l \)th normalized coherency matrix \( \tilde{J}_l \) is given by

\[
\tilde{J}_l = \frac{1}{2} \left( 1 + \tilde{S}_{1,l}^E \tilde{S}_{2,l}^E - i \tilde{S}_{3,l}^E \tilde{S}_{2,l}^E + i \tilde{S}_{3,l}^E \tilde{S}_{1,l}^E \right),
\]
(3.6.6)

thus, by using \( \tilde{J}_l \), Eq. (3.6.5) is written as

\[
\frac{i}{d} \frac{d}{dz} \tilde{J}_l = [\hat{H}', \tilde{J}_l].
\]
(3.6.7)

The left-hand side of Eq. (3.6.7) is written as

\[
\frac{i}{d} \frac{d}{dz} \tilde{J}_l = i \left( \frac{1}{2} \frac{d}{dz} \tilde{S}_l^E \right) \cdot \tilde{\sigma}^{xy}.
\]
(3.6.8)

The right-hand side of Eq. (3.6.7) is calculated as

\[
[\hat{H}', \tilde{J}_l] = \frac{1}{2} [\hat{H}', \sigma_0] + \frac{1}{2} [\hat{H}', (\tilde{S}_l^E) \cdot \tilde{\sigma}^{xy}]
= 0 + \sum_{i=1}^{3} \sum_{j=1}^{3} H_i S_{ij,l}^E \left[ (\tilde{\sigma}^{xy})_i, (\tilde{\sigma}^{xy})_j \right] \frac{1}{2}
= i \left( \hat{H} \times \tilde{S}_l^E \right) \cdot \tilde{\sigma}^{xy}.
\]
(3.6.9)

Here we used the relationship [108] of

\[
[\sigma_i, \sigma_j] = 2i \sum_{k=1}^{3} \epsilon_{ijk} \sigma_k,
\]
(3.6.10)

where \( \epsilon_{ijk} \) is the Levi-Civita symbol. Merging Eqs. (3.6.5), (3.6.8) and (3.6.9), we finally obtain the “Bloch equations” on the \( l \)th EPS

\[
\frac{d}{dz} \tilde{S}_l^E = 2 \hat{H} \times \tilde{S}_l^E,
\]
(3.6.11)
which is the same formula as the precessional motion in classical mechanics [108]; $\hat{H}$ and $\hat{S}_E^l$ are respectively regarded as the torque and the angular momentum. That formula ensures $\hat{S}_E^l$ points on the surface of the $l$th EPS at any $z$.

From Eq. (3.6.11), it is obvious that there are stationary vectors of $\hat{S}_E^l$, which are proportional to $\hat{H}$. The states $\tilde{E}_l^l$ of these vectors can be derived through diagonalization of $\hat{H}'$. When we assume the eigenvectors and the eigenvalues of $\hat{H}'$ are respectively $\lambda_{\pm}$ and $p_{\pm} = (p_{x\pm}, p_{y\pm})^T$, (3.6.12)
is satisfied. That equation is solved as

$$\lambda_+ = +\sqrt{H_2^2 + H_3^2 + H_1^2}, \quad \begin{pmatrix} p_x^+ \\ p_y^+ \end{pmatrix} = \begin{pmatrix} H_1 + \lambda_+ \\ H_2 + iH_3 \end{pmatrix}, \quad (3.6.13)$$

$$\lambda_- = -\sqrt{H_2^2 + H_3^2 + H_1^2}, \quad \begin{pmatrix} p_x^- \\ p_y^- \end{pmatrix} = \begin{pmatrix} -H_2 + iH_3 \\ H_1 - \lambda_- \end{pmatrix}, \quad (3.6.14)$$

where the eigenvectors have the relationship $(p_x^-, p_y^-)^T = (-p_y^+, p_y^+)^T$ [107].

The normalized ESVs $\hat{S}_E^l_{\pm}$ are expressed by

$$\hat{S}_E^l_{\pm} = \pm \frac{\hat{H}}{|H|}, \quad (3.6.15)$$

which corresponds to the same result in the former discussion.

**Jones matrix corresponding to $\hat{H}'$**

We have obtained the “Hamiltonians” representing arbitrarily optic elements that makes the $l$th normalized ESV move on the surface of the $l$th EPS. In this section, we will acquire the corresponding expression of Jones matrices $\hat{O}$ to the “Hamiltonians” $\hat{H}'$.

The Jones matrix $\hat{O}(\equiv R_{l\phi} \hat{O} R_{-l\phi})$ satisfies

$$\hat{E}^\text{out}_\perp = \hat{O} \hat{E}^\text{in}_\perp, \quad (3.6.16)$$

$$\hat{E}^\text{out}_\perp = \hat{O} \hat{E}^\text{lin}_\perp \quad (3.6.17)$$

where $\hat{E}^\text{in}_\perp (= R_{l\phi} \hat{E}^\text{lin}_\perp)$ and $\hat{E}^\text{out}_\perp (= R_{l\phi} \hat{E}^\text{lin}_\perp)$ are respectively the transverse electric vectors at the input facet ($z = 0$) and the output facet ($z = d$) of the

---

2The definition of the Jones matrix is described in Appendix. A.2.
optical element expressed by $\hat{O}$. For simplicity, we consider only the light states with $P_l = P_l^{(\text{space})} = 1$.

When we consider the inside of the optical element ($0 < z < d$), Eq. (3.6.17) is rewritten as

$$\bar{E}_\perp^l(z) = \hat{O}_l(z)\bar{E}_\perp^\text{in}$$

(3.6.18)

where

$$\bar{E}_\perp^l(d) \equiv \bar{E}_\perp^\text{out}, \quad \hat{O}_l(d) \equiv \hat{O}_l.$$  

(3.6.19)

By differentiating the both side of Eq. (3.6.18) by $z$, we acquire

$$\frac{d}{dz}\bar{E}_\perp^l(d) = \left(\frac{d}{dz}\hat{O}_l(z)\right)\bar{E}_\perp^\text{in}$$

$$= \left(\frac{d}{dz}\hat{O}_l(z)\right)\hat{O}_l^\dagger(z)\left(\hat{O}_l(z)\bar{E}_\perp^\text{in}\right)$$

$$= \left[\left(\frac{d}{dz}\hat{O}_l(z)\right)\hat{O}_l^\dagger(z)\right]\bar{E}_\perp^l(d).$$  

(3.6.20)

By comparing this equation with Eq. (3.6.3), we find

$$i\left(\frac{d}{dz}\hat{O}_l(z)\right)\hat{O}_l^\dagger(z) \equiv \hat{H}'.$$

(3.6.21)

Thus we acquire the second order differential equation:

$$\frac{d^2}{dz^2}\hat{O}_l(z) = -(\hat{H}')^2\hat{O}_l(z).$$

(3.6.22)

Since $(\hat{H}')^2 = |\hat{H}|^2 I \equiv \lambda^2 I$ ($I$ is the unit matrix and $\lambda > 0$), that equation is solved as

$$\hat{O}_l(z) = I \cos \lambda z + B \sin \lambda z$$

$$\equiv \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \cos \lambda z + \begin{pmatrix} b_{11} & b_{12} \\ b_{21} & b_{22} \end{pmatrix} \sin \lambda z,$$

(3.6.23)

(3.6.24)

which satisfies the initial condition $\hat{O}_l(0) \equiv I$. Since $\hat{O}_l$ satisfies Eq. (3.6.22), the matrix $B$ is expressed as

$$B = -\frac{i}{\lambda} \begin{pmatrix} H_1 & H_2 - iH_3 \\ H_2 + iH_3 & -H_1 \end{pmatrix}.$$  

(3.6.25)

Finally, we obtain

$$\hat{O}(z) = I \cos \lambda z - \frac{i}{\lambda} R_{l\phi} \begin{pmatrix} H_1 & H_2 - iH_3 \\ H_2 + iH_3 & -H_1 \end{pmatrix} R_{-l\phi} \sin \lambda z.$$  

(3.6.26)
Fast-axis distributions of various $q$-retarders. The values of $(q, \alpha_0)$ are (a) $\left(\frac{1}{2}, 0\right)$, (b) $\left(\frac{1}{2}, \pi/4\right)$, (c) $\left(1, \pi/2\right)$ and (d) $\left(1, 0\right)$ \[127\].

**Hamiltonian for $q$-retarders**

Here, we discuss the “Hamiltonian” for $q$-retarders \[127\]. The $q$-retarder is one of the inhomogeneous anisotropic media. The angle $\alpha$ of the fast axis at $(r, \phi)$ is distributed as

$$\alpha(r, \phi) = q(\phi + \alpha_0),\quad (3.6.27)$$

where $q$ and $\alpha_0$ are respectively the integer or half-integer and arbitrarily angle parameters to specify the fast-axis distribution of $q$-retarders (Fig. 3.6.1). The Jones matrix for the $(q, \alpha_0)$-retarder is expressed by

$$\hat{O}_{\text{qret}}(\delta, q, \alpha_0) = R_{q(\phi + \alpha_0)} \begin{pmatrix} e^{-i\delta/2} & 0 \\ 0 & e^{i\delta/2} \end{pmatrix} R_{-q(\phi + \alpha_0)}$$

$$= I \cos \frac{\delta}{2} - i R_{q\phi} \begin{pmatrix} \cos 2q\alpha_0 & \sin 2q\alpha_0 \\ \sin 2q\alpha_0 & -\cos 2q\alpha_0 \end{pmatrix} R_{-q\phi} \sin \frac{\delta}{2}.\quad (3.6.28)$$

where $\delta$ is retardance of the $(q, \alpha_0)$-retarder. By comparing this Jones matrix with the one in Eq. (3.6.24), we obtain

$$\lambda = 1$$

$$l = q$$

$$z = \frac{\delta}{2\lambda}.$$  \hspace{1cm} (3.6.31)

$$H_1 = \lambda \cos 2q\alpha_0,$$  \hspace{1cm} (3.6.32)
\[ H_2 = \lambda \sin 2q\alpha_0, \quad (3.6.33) \]
\[ H_3 = 0. \quad (3.6.34) \]

From Eq. (3.6.30), we consider only when \( q \) is integer because \( l \) must be integer. Thus, the Hamiltonian for the \((q, \alpha_0)\)-retarder is given by

\[ \hat{H}_q^{\text{ret}} = \cos(2q\alpha_0)\sigma_3 + \sin(2q\alpha_0)\sigma_1. \quad (3.6.35) \]

The “Bloch equations” on the \( q \)th EPS is written by

\[ \frac{d}{d\delta} \tilde{S}_l^E = \begin{pmatrix} \cos 2q\alpha_0 \\ \sin 2q\alpha_0 \\ 0 \end{pmatrix} \times \tilde{S}_l^E, \quad (3.6.36) \]

which means that the torque of the precession \( \hat{H} \) is always on the \((S_1^E, S_2^E)\) plane.

### 3.6.2 Berry connection and Berry phase on an extended Poincaré sphere

We extract the PBP on the EPS through the Berry connection of \( H' \). Since \( H' \) can be expressed by the linear combination of Pauli spin matrices, \( H' \) is classified with spin-1/2 Hamiltonian \([116] \). Consequently, the Berry connection \( A(\tilde{S}_l^E) \) is described by \([128] \)

\[ A(\tilde{S}_l^E) = -e^{\Phi} e_{\theta_l^E} - \frac{1}{2} \cos(\theta_l^E) \tan \frac{\theta_l^E}{2} + \nabla \tilde{S}_l^E \Phi(\tilde{S}_l^E), \quad (3.6.37) \]

where \( \Phi(\tilde{S}_l^E) \) is the gauge function, \((R_l^E, \phi_l^E, \theta_l^E)\) is the spherical coordinate of the \( l \)th EPS and \( e_{R_l^E}, e_{\phi_l^E} \) and \( e_{\theta_l^E} \) are spherical bases defined by

\[ \tilde{S}_l^E = R_l^E e_{R_l^E} + \phi_l^E e_{\phi_l^E} + \theta_l^E e_{\theta_l^E}, \quad (3.6.38) \]

\[ 0 \leq \phi_l^E < 2\pi, \]

We consider a trajectory described by a closed loop \( C \) on the \( l \)th EPS. Hence, the PBP phase \( \gamma_{\text{PBP}} \) is given by \([116] \)

\[ \gamma_{\text{PBP}} = \oint_C A(\tilde{S}_l^E) \cdot d\tilde{S}_l^E = -\frac{\Omega}{2}, \quad (3.6.39) \]

where \( \Omega \) is the solid angle subtended by the contour \( C \) (Fig. 3.6.2).
Figure 3.6.2: The conceptual drawing of the PBP on the $l = 1$ EPS.
Chapter 4

Generation and characterization of cylindrically polarized broadband pulses

In this chapter, we show usefulness of using the ESPs and the DOP-SD through experimental full-quantitative characterization of CP broadband pulses\(^1\). First, we overview the generation method of CP modes (Sec. 4.1) and the concept to generate arbitrarily CP broadband pulse state, which we use in this experiment (Sec. 4.2). After that, experimental setup and results are presented in Sec. 4.3. In order to evaluate the accuracy of the full-quantitative characterization, we compare the experimental results with simulation results (Sec. 4.4). Finally, we discuss the usefulness of full-quantitative characterization of CP broadband pulses by use of the ESPs and the DOP-SD in Sec. 4.5.

4.1 Background

We have reviewed the CP modes and the measurement method for them in Sec. 1. In this section, we overview background from a standpoint of the generation method of the CP modes.

There are basically three ways to generate the CP modes as follows:

- Coherent beam combining [57, 58, 63, 67, 90, 91]
- Direct producing from a resonator [59–62, 64–66, 69, 70, 72–76, 82, 83, 86]

\(^1\)This chapter is partly based on the paper [113] (http://dx.doi.org/10.1038/srep17797)
• Polarization converters [68, 71, 77–81, 84, 85, 88, 89, 129]

Almost all the direct producing methods generate CP continuous wave (CW) beams. In contrast to that, the coherent beam and polarization converter methods can be utilized in order to generate both CP CW and pulse beams. In terms of the number of modes and CP states which can be generated, the coherent beam combining method is superior to the other methods because the direct producing and polarization converter methods are generally $l$-fixed.

In this chapter, we will show the general usefulness of the ESPs and the DOP-SD for characterizing CP states. We therefore use the coherent beam combining method and generate various CP broadband pulse states.

4.2 Concept of generating arbitrarily cylindrically polarized broadband pulse states

We here describe the basic concept of generating arbitrary CP broadband pulses (Fig. 4.2.1(a)). The detail of the experimental setup is shown in Sec. 4.3.1. First, $x$-polarized $|l = 0\rangle$ broadband (or ultrashort) pulses ($E_0$) are converted into $x$-polarized $|l = m, p = 0\rangle$ OV ($E_1$) by the spatial light modulator in the 4-$f$ configuration (4-$f$ SLM). Here, $l$ and $p$ are respectively referred to as the azimuthal and the radial indices of LG modes [46]. A super-achromatic half-wave plate (HWP1) based on the design by Pancharatnam [130] changes the angle of linear polarization ($E_3$). After that, a coherent combining system coherently superposes $x$-polarized $|l = m, p = 0\rangle$ and $y$-polarized $|l = -m, p = 0\rangle$ OV broadband pulses ($E_4$), whose energy ratio is controlled by HWP1; $\cos^2(2\theta_{H1}) : \sin^2(2\theta_{H1})$. After that, the $x$- and $y$-polarized components of $E_4$ are respectively converted into $|s = -1\rangle$ and $|s = +1\rangle$ circularly polarized states $E_5$ by a super-achromatic quarter-wave plate (QWP1). Here, $s$ is the spin angular momentum of photon in units of $\hbar$ [114]. The pulse passes through a super-achromatic half-wave plate (HWP2), following which the sign of spin angular momentum of light is flipped [94]
Figure 4.2.2: The relationship of the rotational angles $\theta_{H1}$ and $\theta_{H2}$ to a generated beam state ((a) $m = 1$ and (b) $m = -1$).

and the relative phase between $|s=+1\rangle$ and $|s=-1\rangle$ states can be adjusted by the rotation angle of HWP2 $\theta_{H2}$:

$$E_6 = \cos(2\theta_{H1})e^{-2i\theta_{H2}}|s=+1\rangle|l=-m, p=0\rangle - i \sin(2\theta_{H1})e^{2i\theta_{H2}}|s=-1\rangle|l=+m, p=0\rangle,$$

which gives $m$th CP broadband pulses [99]. The normalized TSPP ESV of the pulse state is

$$S_m^{E,TSPP} = \begin{pmatrix} -\sin(4\theta_{H1}) \sin(4\theta_{H2}) \\ \sin(4\theta_{H1}) \cos(4\theta_{H2}) \\ \cos(4\theta_{H1}) \end{pmatrix},$$

which is represented by the point $(\theta_m^E, \phi_m^E) = (4\theta_{H1}, \pi/2 + 4\theta_{H2})$ on the TSPP EPS (Fig. 4.2.2). Hence arbitrary manipulation of CP broadband pulse state can be achieved by adjusting the rotation angles of HWP1 and HWP2.

4.3 Experimental

4.3.1 Setup

Figure 4.3.1 shows an experimental setup. This setup is composed of 5 parts; 4-f SLM (generation of broadband optical vortex pulses), coherent combining, CP beam generation, spectral interference and monitoring, and polarization measurement system.
Figure 4.3.1: Setup for generating arbitrary CP broadband pulses by use of coherent beam combining. SLM, a spatial light modulator (available wavelength ranges from 620 nm to 1100 nm); HWP1,2, super-achromatic half-wave plates (from 600 nm to 2700 nm); HWP3, an achromatic half-wave plate (from 690 nm to 1200 nm); PBS, a low-group-velocity-dispersion polarizing beam splitter (from 680 nm to 1080 nm); P1-3, periscopes; QWP1, a super-achromatic quarter-wave plate (from 600 nm to 2700 nm); QWP2, an achromatic quarter-wave plate (from 690 nm to 1200 nm); FM, a flip mirror; POL1,2, polarizers; Sampler, a beam sampler; BPF, a bandpass filter; GLP, a Glan-Laser polarizer; CCD1,2, charge-coupled-device cameras.

**Generation of broadband optical vortex pulses.**

The generated pulses from a Ti:Sa laser amplifier (center wavelength 800 nm, bandwidth of ~40 nm, pulse duration ~25 fs, and repetition rate 1 kHz), whose temporal waveform is represented by \( f(t, z) \), are attenuated by ND filters, following which the 4-\( f \) SLM converts into \( x \)-polarized \( l = 1, p = 0 \) or \( l = 2, p = 0 \) OV pulses. The 4-\( f \) configuration in the spatial light modulator (SLM) system enables us to compensate for angular dispersion [131, 132]. We furthermore utilize a complex-amplitude modulation technique with a phase-only SLM [133–136] as means to convert to broadband arbitrary single LG mode OV pulses. Hence, the transverse electric field \( \mathbf{E}_1 \) is described as

\[
\mathbf{E}_1 = f(t, z)g(x, y, z)e^{i\phi} \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad (4.3.1)
\]

\[
\hat{S}_{0, \mathbf{E}_1}^{E, \text{TSPP}} = (1, 0, 0), \quad (4.3.2)
\]
Figure 4.3.2: Trajectories on the $l = 0$ TSPP ESPs. (a) The trajectory from $E_0$ to $E_3$ when $\theta_{H1} = -\pi/8$. (b) The trajectories of magenta and blue components from $E_3$ to $E_0$.

where $g$ represents the amplitude profile ideally described by

$$g(x, y, z) = u_{m,0}^{\text{CPLG}}(x, y, z),$$  \hspace{0.5cm} (4.3.3)

and $S_{0, E}^\text{E.TSPP}$ means that the 0th TSPP ESPs of $E$.

**Coherent combining system**

After passing through a super-achromatic half-wave plate (HWP1), the transverse electric field $E_2$ is represented by

$$E_2 = f(t, z)g(x, y, z) \begin{pmatrix} \cos(2\theta_{H1}) \\ -\sin(2\theta_{H1}) \end{pmatrix} e^{im\phi},$$  \hspace{0.5cm} (4.3.4)

$$S_{0, E_2}^\text{E.TSPP} = (\cos(4\theta_{H1}), -\sin(4\theta_{H1}), 0).$$  \hspace{0.5cm} (4.3.5)

The direction of pulses is changed by 90° by a periscope (P1) whose two mirrors are placed with a twist. The periscope swaps the electric components $E_x$ and $E_y$. The Jones vector thereby is rewritten as follows:

$$E_3 = f(t, z)g(-x, -y, z) \begin{pmatrix} -\sin(2\theta_{H1}) \\ \cos(2\theta_{H1}) \end{pmatrix} e^{im\phi},$$  \hspace{0.5cm} (4.3.6)

$$S_{0, E_3}^\text{E.TSPP} = (-\cos(4\theta_{H1}), -\sin(4\theta_{H1}), 0).$$  \hspace{0.5cm} (4.3.7)

Trajectory on the $l = 0$ TSPP ESPs from $E_0$ to $E_3$ is described by Fig. 4.3.2.
The pulse is divided into two by a low-group-velocity-dispersion polarization beam splitter (PBS). The numbers of reflection in the blue branch and the magenta branch in Fig. 4.3.1 are 6 and 7, respectively. Thus, the beam cross-sectional profile of the pulses in the magenta branch is flipped \((g(-x, -y, z)e^{im\phi} \rightarrow g(x, -y, z)e^{-im\phi})\). An achromatic half-wave plate (HWP2) makes a polarization rotation of pulses in the both blue and magenta branches by 90°. The \(x\)-polarized \(|l = -m, p = 0\rangle\) and \(y\)-polarized \(|l = m, p = 0\rangle\) OVs are hence coherently combined at PBS:

\[
E_4 = g(-x, -y, z) \begin{pmatrix} f(t + \tau, z) \cos(2\theta_{H1})e^{-im\phi} \\ f(t, z) \sin(2\theta_{H1})e^{im\phi} \end{pmatrix} = E_4^{\text{Magenta}} + E_4^{\text{Blue}},
\]

(4.3.8)

\[
\hat{S}_{0,E_4^{\text{Magenta}}} = (1, 0, 0),
\]

(4.3.9)

\[
\hat{S}_{0,E_4^{\text{Blue}}} = (-1, 0, 0),
\]

(4.3.10)

where \(E_4^{\text{Magenta}}\) and \(E_4^{\text{Blue}}\) are respectively the electric components of the magenta and the blue branches described as

\[
E_4^{\text{Magenta}} = g(-x, -y, z)f(t + \tau, z) \cos(2\theta_{H1})e^{-im\phi} \begin{pmatrix} 1 \\ 0 \end{pmatrix},
\]

(4.3.11)

\[
E_4^{\text{Blue}} = g(-x, -y, z)f(t, z) \sin(2\theta_{H1})e^{im\phi} \begin{pmatrix} 0 \\ 1 \end{pmatrix}.
\]

(4.3.12)

Here \(\tau\) is delay time between pulses in the blue and magenta branches, which is controlled by the piezo driver to be \(\tau = 0\). We assume that the amplitude profile \(g\) has a bilaterally symmetric \(g(x, -y) = g(-x, -y)\) since OV has doughnut-shaped intensity profile.

Cylindrically polarized mode conversion

A super-achromatic quarter-wave plate (QWP1) converts the polarization states of the electric components of the magenta and the blue branches into circularly polarized:

\[
\begin{align*}
E_5 &= \left[ E_4^{\text{Blue}} \cdot \begin{pmatrix} 0 \\ 1 \end{pmatrix} \right] \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix} + \left[ E_4^{\text{Magenta}} \cdot \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right] \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -1 \end{pmatrix} \\
&= E_5^{\text{Blue}} + E_5^{\text{Magenta}},
\end{align*}
\]

\[
\begin{align*}
&= g(-x, -y, z) \left[ f(t + \tau, z) \cos(2\theta_{H1})e^{-m} + f(t, z) \sin(2\theta_{H1})e^{m} \right] \\
&\approx E_5^{\text{Blue}} + \hat{S}_{0,E_5^{\text{Magenta}}} = (0, 0, -1),
\end{align*}
\]

(4.3.13)

(4.3.14)
Using a super-achromatic half-wave plate (HWP1), we can adjust the relative phase between the electric components of the magenta and the blue branches.

\[
E_6 = \begin{bmatrix} E_4^\text{Magenta} \cdot (1 \ 0)^\top \frac{e^{-2i\theta_{h2}}}{\sqrt{2}} \ (1 \ i) \\ E_4^\text{Blue} \cdot (0 \ 1)^\top \frac{ie^{2i\theta_{h2}}}{\sqrt{2}} \ (1 \ -i) \end{bmatrix} \equiv E_6^\text{Magenta} + E_6^\text{Blue},
\]

\[
E_6^\text{Magenta} = g(-x, -y, z)[f(t, z) \sin(2\theta_{h1})e^{-2i\theta_{h2}}e_+^m - if(t + \tau, z) \cos(2\theta_{h1})e^{2i\theta_{h2}}e_-^m],
\]

\[
S_{E,TSPP}^{E,TSPP} = (0, 0, 1), \quad (4.3.16)
\]

\[
S_{E,TSPP}^{E,TSPP} = (0, 0, -1). \quad (4.3.17)
\]

\[
S_{E,TSPP}^{E,TSPP} = (0, 0, 1). \quad (4.3.18)
\]

Finding the zero delay by using the spectral interference technique

Using a polarizer (POL2) and a spectrometer, we find the zero delay with the aid of the spectrum interference method \[137\]. A charge-coupled-device (CCD1) monitors the intensity profile of the \(x\)-polarized component of \(E_5\), namely,

\[
|E_5 \cdot (1, 0)^\top|^2 \simeq 2g(-x, -y, z)f(t, z)^2 \left[ 1 + \cos \left( m\phi - \frac{\omega}{2}\tau \right) \right], \quad (4.3.19)
\]

in order to ensure the delay time is unchanged within the polarization measurement.

Finally, the arbitrarily CPLG pulses are generated. The transverse electric component \(E_6\) and TSPP ESV \(\tilde{S}_{E,TSPP}^{E,TSPP}\) are represented by

\[
E_6 = g(-x, -y, z)f(t, z) \left[ \cos(2\theta_{h1})e^{-2i\theta_{h2}}e_+^m - i\sin(2\theta_{h1})e^{2i\theta_{h2}}e_-^m \right], \quad (4.3.20)
\]

and

\[
\tilde{S}_{E,TSPP}^{E,TSPP} = \begin{bmatrix} -\sin(4\theta_{h1}) \sin(4\theta_{h2}) \\ \sin(4\theta_{h1}) \cos(4\theta_{h2}) \\ \cos(4\theta_{h1}) \end{bmatrix}. \quad (4.3.21)
\]

Measuring polarization distributions

In the polarization measurement system, the pulses are spectrally-resolved by bandpass filters (BPF; center wavelengths, 780, 790, 800, 810, 820 nm; bandwidths, 10 nm), then their polarization distributions are acquired by using
a rotating-retarder type imaging polarimeter [110], which is composed of an achromatic quarter-wave plate (QWP2), a Glan-Laser polarizer (GLP) and a charge-coupled-device camera (CCD2). From the polarization distribution, we computed the normalized TSPP ESV $\tilde{S}_{E,TSPP}^m$ and the $m$th DOP-SD $\mathcal{P}_{l=m}^{\text{space}}$. Here, the origins $(x,y) = (0,0)$ on the recorded images are set to maximize the $m$th DOP-SD.

4.3.2 Results

We respectively generated seven states for $l = 1$ and $l = 2$ CP broadband pulses: $(\theta_{E_m}^E, \phi_{E_m}^E) = (0,0), (\pi/4,0), (\pi/4,\pi/4), (\pi/4,\pi/2), (\pi/2,0), (\pi/2,\pi/4), (\pi/2,\pi/2)$. For simplicity, $(\theta_{E_m}^E, \phi_{E_m}^E)$ is omitted hereafter. Figure 4.3.3 shows characterization results for $l = 1 (\pi/2,0)$ and $l = 2 (\pi/2,0)$ CP pulses as typical examples. Spectrally-resolved polarization distributions are shown in Figs. 4.3.3(a) and 4.3.3(d); (a) is for $l = 1 (\pi/2,0)$ CP pulses (or radially polarized pulses) and (d) is for $l = 2 (\pi/2,0)$ CP pulses. From the polarization distributions in Figs. 4.3.3(a) and 4.3.3(d), the values of $\tilde{S}_{E,TSPP}^{1,1}$ (Fig. 4.3.3(b)) and $\mathcal{P}_{1}^{\text{space}}$ (Fig. 4.3.3(c)), and $\tilde{S}_{E,TSPP}^{2,1}$ (Fig. 4.3.3(e)) and $\mathcal{P}_{2}^{\text{space}}$ (Fig. 4.3.3(f)) in individual spectral ranges were computed.

The characterization results for all states are described in Figs. 4.3.4; (a) and (b) are for $l = 1$ CP pulse states and (c) and (d) are for $l = 2$ CP pulse states. Figures 4.3.4(a) and 4.3.4(c) respectively represent the $l = 1$ and $l = 2$ TSPP EPSs, on which the spectrally-resolved values of normalized ESVs $\tilde{S}_{E,TSPP}^{l=1,2}$ in $l = 1$ and $l = 2$ CP states are plotted. The spectrally-resolved values of DOP-SD corresponding to the CP states in Figs. 4.3.4(a) and 4.3.4(c) are shown in Figs. 4.3.4(b) and 4.3.4(d), respectively.
Figure 4.3.3: Characterization results of $l = 1$ ((a), (b) and (c)) and $l = 2$ ((d), (e) and (f)) $(\pi/2,0)$ CP broadband pulses. (a) and (d) spectrally-resolved polarization distribution of generated $l = 1$ and $l = 2$ CP pulses, respectively. These polarization distributions are colored under the following rule: red, left-handed elliptical polarization; blue, right-handed elliptical polarization; black, linear polarization. The green points at the center of images represent the origins $(x,y) = (0,0)$. (b) and (c) characterization results of $S_{1,1}^E$ and $P_{1}\text{space}^{\text{E}}$ for $l = 1$ $(\pi/2,0)$ CP pulses, respectively. (e) and (f) characterization results of $S_{1,2}^E$ and $P_{2}\text{space}^{\text{E}}$ for $l = 2$ $(\pi/2,0)$ CP pulses, respectively.
Figure 4.3.4: Spectrally-resolved characterization results for \( l = 1 \) ((a) and (b)) and \( l = 2 \) ((c) and (d)) CP pulses. The seven CP pulse states are realized in every azimuthal index \( l \). (a) and (c) The values of TSPP ESV \( \tilde{S}^{E}_{l,TSSP} \) for \( l = 1 \) and \( l = 2 \) CP pulse states plotted on the TSPP EPS, respectively. (b) and (d) DOP-SD \( P^{l}_{SD} \) of \( l = 1 \) and \( l = 2 \) CP pulse states corresponding to (a) and (c), respectively.
4.4 Simulation

Figures 4.4.1(a), 4.4.1(b), 4.4.1(c) and 4.4.1(d) respectively depict the intensity and polarization distributions of $(l, \theta_H) = (1, 0)$, $(1, \pi/4)$, $(2, 0)$ and $(2, \pi/4)$ cases. The measurements of Figs. 4.4.1(a) and 4.4.1(c), and 4.4.1(b) and 4.4.1(d) are respectively conducted under blocking the blue branch and the magenta branch in Fig. 4.3.1, which means that $E_6$ should be the linear combination of $|s = +1\rangle|l = -m\rangle$ and $|s = -1\rangle|l = +m\rangle$. However, these intensity distributions are of twofold symmetry rather than axisymmetry. This result is attributed to the slight superimposition of $|l = m \pm 2\rangle$ component on $|l = m\rangle$ OV pulses because of deformation passing through optic elements. Though the polarization distribution should be circularly polarized, the polarization states are elliptic. This fact can be ascribed to the retardation errors of super-achromatic wave plates. The actual electric field of $E_6$ is approximately described as

$$E_6 = e^{-2i\theta_{H2}}|s = +1\rangle \left[ \cos 2\theta_{H1}|l = -m\rangle + \delta_1|l = -m + 2\rangle + \delta_2|l = -m - 2\rangle \right]$$

$$-i \sin 2\theta_{H1}\delta_3|l = +m\rangle$$

$$+ e^{2i\theta_{H2}}|s = -1\rangle \left[ -i \sin 2\theta_{H1}|l = +m\rangle + \delta_4|l = +m - 2\rangle + \delta_5|l = +m + 2\rangle \right]$$

$$+ \cos 2\theta_{H1}\delta_6|l = -m\rangle],$$

where $\delta_{1,2,4,5}$ and $\delta_{3,6}$ are superposition coefficients associated with the deformation and the elliptical polarization, respectively. When $m = 1$, the individual unwanted terms

$$e^{-2i\theta_{H2}}(\cos 2\theta_{H1}\delta_1 - i \sin 2\theta_{H1}\delta_3)|s = +1\rangle|l = 1\rangle$$

and

$$e^{2i\theta_{H2}}(-i \sin 2\theta_{H1}\delta_4 + \cos 2\theta_{H1}\delta_6)|s = -1\rangle|l = -1\rangle$$

can be partly canceled. However, in the $m = 2$ case, the unwanted terms are

$$e^{-2i\theta_{H2}}|s = +1\rangle(\cos 2\theta_{H1}(\delta_1|l = 0\rangle + \delta_2|l = 4\rangle) - i \sin 2\theta_{H1}\delta_3|l = 2\rangle$$

and

$$e^{2i\theta_{H2}}|s = -1\rangle(-i \sin 2\theta_{H1}(\delta_4|l = 0\rangle + \delta_5|l = 4\rangle) + \cos 2\theta_{H1}\delta_6|l = 2\rangle),$$

which cannot be canceled. The contamination of terms except $|s = +1\rangle|l = -m\rangle$ and $|s = -1\rangle|l = +m\rangle$ leads to degradation of $C_{|m-1\rangle}$ rotational symmetry. The value of DOP-SD of $l = 2$ CP pulses are thus smaller than that of $l = 1$ pulses.
Figure 4.4.1: (a)-(d) The intensity and polarization distributions of \((l, \theta_{H1}) = (a) (1, 0), (b) (1, \pi/4), (c) (2, 0) and (d) (2, \pi/4)\) cases. The experimental measurements of (a) and (c), and (b) and (d) are conducted under blocking the blue branch and the magenta branch in Fig. 4.3.1, respectively. The white and black arrows are placed to emphasize the twofold symmetry of the intensity patterns. All the polarization distributions are colored under the rule in Fig. 4.3.3.

Table 4.4.1: Evaluated values of \(\delta_{1,2,3,4,5,6}\)

<table>
<thead>
<tr>
<th>(l)</th>
<th>(\delta_1)</th>
<th>(\delta_2)</th>
<th>(\delta_3)</th>
<th>(\delta_4)</th>
<th>(\delta_5)</th>
<th>(\delta_6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(-0.08e^{2.20i})</td>
<td>0</td>
<td>0.14e^{-2.32i}</td>
<td>0.08e^{-0.34i}</td>
<td>0</td>
<td>0.15e^{-0.44i}</td>
</tr>
<tr>
<td>2</td>
<td>0.08e^{-0.30i}</td>
<td>0</td>
<td>0.14e^{-2.52i}</td>
<td>0.08e^{-0.34i}</td>
<td>0</td>
<td>0.15e^{-0.44i}</td>
</tr>
</tbody>
</table>

We conducted simulation for \(l = 1 (\pi/2, 0)\) and \(l = 2 (\pi/2, 0)\) CP states. First, we respectively evaluated \(\delta_{1,2,4,5}\) and \(\delta_{3,6}\) from the intensity and the polarization distributions in Figs. 4.4.1(a), 4.4.1(b), 4.4.1(c) and 4.4.1(d) (the values are in Table 4.4.1). The simulation results are shown in Figs. 4.4.2(a), 4.4.2(b) and Table 4.4.2. The intensity distributions were plotted by using the following equation based on Eq. (4.4.1):

\[
|\mathbf{E}_6'(r, \phi)|^2 \propto \left[|\cos 2\theta_{H1}(e^{-im\phi} + \delta_1 e^{-(m+2)\phi} + \delta_2 e^{-(m-2)\phi}) - i \sin 2\theta_{H1}\delta_3 e^{im\phi}|^2 \\
+ |\sin 2\theta_{H1}(e^{im\phi} + \delta_4 e^{(m-2)\phi} + \delta_5 e^{(m+2)\phi}) + \cos 2\theta_{H1}\delta_6 e^{-im\phi}|^2\right] \\
r^{2|m|} \exp \left(-\frac{2r^2}{w^2}\right),
\]

(4.4.2)

where \((r, \phi)\) is the circular polar coordinates and \(w\) is a parameter for the beam size. We have made simulations under the various conditions of \(\delta_{1,2,4,5}\), and confirmed that the values of \(\tilde{S}_{l,l}^F\) and \(\mathcal{P}_{l,\text{space}}\) hardly changed.
Figure 4.4.2: The simulation results for (a) \(l = 1 (\pi/2, 0)\) and (b) \(l = 2 (\pi/2, 0)\) pulse states. All the polarization distributions are colored under the rule in Fig. 4.3.3.

Table 4.4.2: Simulation conditions and values of \(\tilde{S}_{l,l}^{E}\) and DOP-SD \(P_{1}^{\text{space}}\).

<table>
<thead>
<tr>
<th>(l = m)</th>
<th>(\theta_{H_1})</th>
<th>(\theta_{H_2})</th>
<th>(\tilde{S}_{1,l}^{E,\text{TSPP}})</th>
<th>(\tilde{S}_{2,l}^{E,\text{TSPP}})</th>
<th>(\tilde{S}_{3,l}^{E,\text{TSPP}})</th>
<th>(P_{l}^{\text{space}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(-\pi/8)</td>
<td>(\pi/8)</td>
<td>0.999</td>
<td>-0.008</td>
<td>-0.015</td>
<td>0.989</td>
</tr>
<tr>
<td>2</td>
<td>(-\pi/8)</td>
<td>(\pi/8)</td>
<td>1.000</td>
<td>0.000</td>
<td>0.000</td>
<td>0.973</td>
</tr>
</tbody>
</table>

### 4.5 Discussion

All polarization distributions of \(l = 1 (\pi/2, 0)\) CP pulses at measured wavelengths \((780, 790, 800, 810\) and \(820\) nm) in Fig. 4.3.3(a) are almost purely RP. This fact is well indicated by the obtained results that \(\tilde{S}_{l,l}^{E}\) and \(P_{1}^{\text{space}}\) were respectively over 0.99 and 0.98 in all spectral regions (Figs. 4.3.3(b) and 4.3.3(c)). Since \(\tilde{S}_{l,l}^{E}\) is associated with the energy ratio between \((\pi/2, 0)\) (RP) state and \((\pi/2, \pi)\) (AP) state [114], which is given by

\[
\frac{\tilde{S}_{l,l}^{E} + 1}{2} : \frac{\tilde{S}_{l,l}^{E} - 1}{2},
\]

over 99% energy of the TSPP state was radially polarized. Moreover, DOP-SD \(P_{l}^{\text{space}}\) enables us to evaluate the over 98% of the temporally-perfect-polarized state of the generated pulses were TSPP state. Consequently, the pulses generated from a coherent combining system had high purity of \(l = 1 (\pi/2, 0)\) CP state and high symmetry of polarization distribution.

From Figs. 4.3.3(e) and 4.3.3(f), \(l = 2 (\pi/2, 0)\) CP pulses similarly had high purity (over 99% in energy ratio) of \(l = 2 (\pi/2, 0)\) CP state and high symmetry (around 97% in energy ratio) in \(l = 2\) CP state, though \(l = 2\) pulses were slightly inferior to \(l = 1\) pulses with regard to symmetry. Contamination of elliptical polarization in the polarization distribution (Fig. 4.3.3(d)) apparently affects the degradation in DOP-SD compared to that of \(l = 1\)
As is mentioned in Sec. 4.4, the contamination comes from two factors. One is the deformation of incident OV pulses into the coherent combining system; the other is the retardation error of super-achromatic wave plates. Both calculated intensity and polarization distributions in Figs. 4.4.2(a) and 4.4.2(b) well agree with that of the experimental results for \( l = 1 (\pi/2, 0) \) (Fig. 4.3.3(a)) and \( l = 2 (\pi/2, 0) \) (Fig. 4.3.3(d)) states, respectively. The values of \( \tilde{S}^E_{1,l} \) and DOP-SD \( \mathcal{P}^\text{space}_{l} \) in Table 4.4.2 are also in good agreement with the experimental results in Figs. 4.3.3(b) and 4.3.3(e), and 4.3.3(c) and 4.3.3(f), respectively. In particular, there is a small (\( \sim 0.02 \)) difference between \( l = 1 \) and \( l = 2 \) cases in the simulation results for DOP-SD, which also appears in the experimental results. Therefore, it should be stressed that our measurement method is able to detect such small asymmetry.

Figures 4.3.4(a) and 4.3.4(c) respectively indicate the spectral dependence of polarization states of \( l = 1 \) and \( l = 2 \) CP pulses. All the pulse states have quite low spectral dependences thanks to optics for broadband pulses such as super-achromatic wave plates and a low-group-velocity-dispersion polarizing beam splitter. All the values of DOP-SD for \( l = 1 \) and \( l = 2 \) CP pulses have low spectral dependence (\( \Delta \mathcal{P}_{l=1,2} \lesssim 0.01 \)), while the DOP-SD values for \( l = 2 \) CP pulses are somewhat less than those for \( l = 1 \) CP pulses by 0.02 to 0.03 (Figs. 4.3.4(b) and 4.3.4(d)) because of the previously described reasons. These results clearly show that our system employing coherent beam combining is able to generate arbitrary CP broadband pulse states with high symmetry and low spectral dependence, which is fully-quantitatively investigated by ESPs and DOP-SD with high precision.

Frequency chirp compensation can be easily achieved because of optics components for broadband pulses in this system. Characterization results in Figs. 4.3.4(a) and 4.3.4(c) show the dispersions of spectrally-resolved polarization states in individual CP pulse states are small (\( \lesssim 0.05 \) in propagation distance on the TSPP ESPs). CP ultrashort pulses with steady polarization state in the pulse duration, which is especially important for applications for magneto-optical storage [138] and nonlinear spectroscopic polarimetry [51], can be therefore generated with this coherent combining system. This experimental setup, where the accessible spectral range covers the region from 690 nm to 1080 nm (limited by the polarizing beam splitter and half-wave plates), offers us the capability of generating ultrashort CP pulses below 10 fs without polarization distribution dispersion. Moreover, by insertion of a femtosecond polarization pulse shaper [139–141] after the 4-\( f \) SLM system, in place of HWP1 and HWP2, our experimental setup will be able to generate the CP pulses with arbitrary control of temporal CP states on one EPS. Although the issue of fully-spatiotemporal characterization method for
ultrashort pulses with nonuniform polarization distribution still remains, our measurement method is quite useful for precise characterization of ultrashort pulses.

In this coherent combining system, we can generate not only CPLG pulses but also CP Bessel-Gauss pulses [79] and bilaterally symmetric vector vortex pulses. An example of bilaterally symmetric vector vortex pulses is Mathieu vector vortex pulses. Mathieu modes, being diffraction free [142], can be generated by a SLM [143, 144]. Some of them are similar to Bose-Einstein condensation (BEC) states having many vortex filaments [145]. Nonlinear propagation or interaction of Mathieu vector vortex pulses in Kerr media obeys the nonlinear Schrödinger equation is therefore of interest.
Chapter 5

Nonlinear propagation of axisymmetrically polarized pulses in an axisymmetrical system

In order to show that the ESPs and the DOP-SD are useful for nonlinear spectroscopic polarimetry, we investigate the nonlinear propagation of AxP pulses in a uniaxial crystal. First, we make an overview of earlier studies in Sec. 5.1. Then, we theoretically analyze the linear propagation of AxP beam in a uniaxial crystal, which is the base of the nonlinear propagation (Sec. 5.2). After that, we obtain the wave equation of the nonlinear propagation in a uniaxial crystal in Sec. 5.3, following which we show the experimental setup and results (Sec. 5.4), and corresponding simulation results (Sec. 5.5). In Sec. 5.6, we discuss the characterization of the nonlinear propagation of AxP pulses in an axisymmetrical system through the ESPs and the DOP-SD.

5.1 Introduction of propagation of light wave in a uniaxial crystal

Uniaxial crystals have been widely used in linear optics (e.g. wave plates) and nonlinear optics (e.g. frequency conversion). However, the optic axes of these crystal are not basically parallel to the beam axes. When we set the optic axis of a uniaxial crystal along the beam axis, the beam passes through the crystal with $C_\infty$ symmetry. In that configuration, an interesting phenomenon, namely the optical spin-to-orbital angular momentum conversion [146–148], beautifully occurs.
Figure 5.1.1: Linear propagation of an AxP beam in a uniaxial crystal along its optic axis. The symmetry of the beam keeps $C_\infty$ in all the propagation positions because the uniaxial crystal has $C_\infty$ symmetry of the permittivity tensor when the beam propagates along its beam axis.

Figure 5.1.1 shows one of the simplest examples of the spin-to-orbital angular momentum conversion in a uniaxial crystals. The input beam is the $|s=1\rangle|l=-1\rangle$ OV, which is the superposition of RP and AP modes. Since in the uniaxial crystal, the RP and AP modes respectively become the TM and TE modes, the resultant beam is the superposition of RP and AP modes with different beam sizes [114, 149]. Hence, the resultant beam has the $|s=-1\rangle$ component as well as the $|s=1\rangle$ component, which means that the helicity conversion of photon (or the optical spin angular momentum (SAM) conversion) $|s=1\rangle \leftrightarrow |s=-1\rangle$ occurs in the uniaxial crystal. In order to satisfy the conservation of total angular momentum (TAM) $(s+l)\hbar$ of photon [150], the $|s=-1\rangle$ component has the topological charge (or the orbital angular momentum (OAM) of photon) of $l=1$; $|s=-1\rangle|l=1\rangle$ OV is generated.

The symmetry of the beam keeps $C_\infty$ while the beam propagates in the uniaxial crystal. The ESPs and DOP-SD are suitable for analyzing the dynamics of the polarization state in the propagation. In particular, some uniaxial crystals also have $C_\infty$ symmetry of the third-order nonlinear opti-
cal susceptibility. We therefore investigate the nonlinear propagation of AxP pulses and analyze it by using ESPs and DOP-SD, which show potential that ESPs and DOP-SD are useful for exploration of nonlinear effects such as nonlinear spectroscopic polarimetry experiments employing CP pulses [51].

5.2 Linear propagation of axisymmetrically polarized beam in a uniaxial crystal

In this section, we discuss the AxP beam linearly propagating along the optic axis of a uniaxial crystal. This discussion is the foundation of the nonlinear propagation of AxP pulses. We obtain the paraxial linear wave equation for that (Sec. 5.2.1) and analyze the dynamics of the polarization distribution through the ESPs and DOP-SD (Sec. 5.2.2).

5.2.1 Paraxial linear wave equation

We start from Maxwell equations of a dielectric substance [151, 152]:

\[ \nabla \cdot B = 0, \quad (5.2.1) \]
\[ \nabla \times E + \frac{\partial B}{\partial t} = 0, \quad (5.2.2) \]
\[ \nabla \cdot D = 0, \quad (5.2.3) \]
\[ \nabla \times H - \frac{\partial D}{\partial t} = 0, \quad (5.2.4) \]

where \( D, B \) and \( H \) are respectively the electric flux density, the magnetic flux density and the magnetic field. The electric flux density \( D \), the magnetic flux density \( B \) are described by constitutive equations:

\[ D = \epsilon \epsilon_0 E, \quad (5.2.5) \]
\[ B = \mu \mu_0 H, \quad (5.2.6) \]

where \( \epsilon, \mu, \epsilon_0, \) and \( \mu_0 \) are respectively the relative permittivity tensor, the relative permeability tensor, the permittivity of vacuum, and the permeability of vacuum.

We consider a uniaxial crystal whose optic axis is parallel to the propagation axis of the beam. Thus, the relative permittivity tensor is written as

\[ \epsilon = \begin{pmatrix} \epsilon_o & 0 & 0 \\
0 & \epsilon_o & 0 \\
0 & 0 & \epsilon_e \end{pmatrix}, \quad (5.2.7) \]
where $\epsilon_o$ and $\epsilon_e$ are respectively the ordinary and the extraordinary permitivities. We furthermore assume $\mu = 1$.

By merging Eqs. (5.2.1) - (5.2.6), we obtain the wave equation:

$$\nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} + \epsilon \epsilon_0 \partial^2 \frac{\partial}{\partial t^2} \mathbf{E} = 0.$$  \hspace{1cm} (5.2.8)

From Eq. (5.2.3), we obtain the relationship as follows:

$$\nabla_\perp \cdot \mathbf{E}_\perp = \partial_x E_x + \partial_y E_y = -\frac{\epsilon_e}{\epsilon_o} \partial_z E_z,$$  \hspace{1cm} (5.2.9)

where $\nabla_\perp = (\partial_x, \partial_y)^T$. Thus, the wave equation (Eq. (5.2.8)) is transformed into two independent wave equations:

$$(\nabla_\perp^2 + \partial_z^2 + k\epsilon) \left( \frac{\hat{E}_\perp}{\hat{E}_z} \right) = \left( 1 - \frac{\epsilon_o}{\epsilon_e} \right) \left( \nabla_\perp(\nabla_\perp \cdot \hat{E}_\perp) \right).$$  \hspace{1cm} (5.2.10)

Here, we consider (quasi-)monochromatic light wave described as

$$\mathbf{E} = \hat{E}(r, \phi, z) \exp\{i(kz - \omega t)\}$$  \hspace{1cm} (5.2.11)

where $k = \omega/c$ is propagation constant, and $c = 1/\sqrt{\epsilon_o \mu_0}$ is the velocity of light in vacuum. We furthermore make use of the slowly varying envelope approximation ($|\partial_z \hat{E}| \gg |\partial^2_z \hat{E}|$) and finally obtain the paraxial wave equation for the transverse electric fields [153]:

$$\nabla_\perp^2 \hat{E}_\perp + 2ik\sqrt{\epsilon_o} \partial_z \hat{E}_\perp = \alpha \nabla_\perp(\nabla_\perp \cdot \hat{E}_\perp),$$  \hspace{1cm} (5.2.12)

where $\alpha (= 1 - \epsilon_o/\epsilon_e)$ is a parameter to describe anisotropy of the medium, and the paraxial wave equation for $\hat{E}_z$ is neglected because $\hat{E}_z$ is enough small to be negligible under the paraxial approximation.

### 5.2.2 Solution to paraxial linear wave equation

We calculate the solution of AxP modes to the paraxial linear wave equation (Eq. (5.2.12)). In order to simplify the wave equation, we introduce the AxP basis:

$$\hat{E}(r, \phi, z) = \hat{E}_r(r, z)e^1_r + \hat{E}_\phi(r, z)e^1_\phi.$$  \hspace{1cm} (5.2.13)

By use of the AxP representation of the transverse electric vector, Eq. (5.2.12) is rewritten [114, 149, 154–157] as

$$\begin{pmatrix} 2ik\sqrt{\epsilon_o/\epsilon_e} \partial_r \hat{E}_r(r, z) \\ 2ik\sqrt{\epsilon_o} \partial_z \hat{E}_\phi(r, z) \end{pmatrix} = \begin{pmatrix} \partial_r^2 + \frac{1}{r^2} \partial_r - \frac{1}{r^2} \\ 0 \end{pmatrix} \begin{pmatrix} \hat{E}_r(r, z) \\ \hat{E}_\phi(r, z) \end{pmatrix}.$$  \hspace{1cm} (5.2.14)
These equations are the same as the paraxial Helmholtz equation (Eq. (2.1.6)). Thus, the solution is described [149, 156, 157] as

\[
\tilde{E}(r, \phi, z) = \sum_p \left[ u_{1,p,r}^{\text{CPLG}} \left( r, \phi, \sqrt{\frac{\epsilon_o}{\epsilon_r}} z \right) \tilde{u}_{1,p}^r e_r^1 + u_{1,p,\phi}^{\text{CPLG}} \left( r, \phi, \frac{1}{\sqrt{\epsilon_o}} z \right) \tilde{u}_{1,p}^\phi e_\phi^1 \right],
\]

(5.2.15)

where

\[
u_{1,p,i}^{\text{CPLG}}(r, \phi, z) = \sqrt{\frac{2p!}{\pi (p+1)!}} \frac{\sqrt{2r}}{w} L_p^1 \left( \frac{2r^2}{w^2} \right) \frac{w_0}{w} \]
\[
\times \exp \left[ -\frac{r^2}{w^2} + i \left( k_i r^2 \frac{1}{2R} - \psi_{\text{Gouy}}(z) \right) \right] \quad (i = r, \phi),
\]

(5.2.16)

\[
k_r = k \sqrt{\frac{\epsilon_r^2}{\epsilon_o}},
\]

(5.2.17)

\[
k_\phi = k \sqrt{\epsilon_o}.
\]

(5.2.18)

The solution indicates that the RP and the AP modes independently propagate with the Rayleigh lengths \( z_r = \sqrt{\epsilon_r^2 / \epsilon_o z_0} \) and \( z_\phi = \sqrt{\epsilon_o z_0} \), respectively.

We here consider the beam propagating in vacuum (\( z < 0 \)) incidents a uniaxial crystal at \( z = 0 \). When the transverse electric field at \( z = 0 \) is described as

\[
\tilde{E}(r, \phi, z = 0) = \sum_p u_{1,p}^{\text{CPLG}} (r, \phi, z_{\text{in}}) \left( \tilde{u}_{1,p}^r e_r^1 + \tilde{u}_{1,p}^\phi e_\phi^1 \right),
\]

(5.2.19)

the transverse electric field after \( z = 0 \) is as follows [114]:

\[
\tilde{E}(r, \phi, z \geq 0) = \sum_p \left[ u_{1,p,r}^{\text{CPLG}} \left( r, \phi, \sqrt{\frac{\epsilon_o}{\epsilon_r}} z + z_{\text{in}} \right) \tilde{u}_{1,p}^r e_r^1 + u_{1,p,\phi}^{\text{CPLG}} \left( r, \phi, \frac{1}{\sqrt{\epsilon_o}} z + z_{\text{in}} \right) \tilde{u}_{1,p}^\phi e_\phi^1 \right].
\]

(5.2.20)

The normalized ESPs of Eq. (5.2.20) are given by

\[
\tilde{S}_{1,1}^E = \frac{|\tilde{u}_{1,p}^r|^2}{|\tilde{u}_{1,p}^r|^2 + |\tilde{u}_{1,p}^\phi|^2} \quad \tilde{S}_{1,1}^E = \sqrt{1 - (\tilde{S}_{1,1}^E)^2} \cdot \frac{4(4 - \tilde{z}^2) \cos \delta_{r\phi} + 16 \tilde{z} \sin \delta_{r\phi}}{16 + 8\tilde{z}^2 + \tilde{z}^4},
\]

(5.2.21)

(5.2.22)
The trajectories of the normalized ESVs are drawn in Fig. 5.2.1. They depend on the $\tilde{S}_{1,1}^E$ and $\delta_{r\phi}$, but are independent of $z$. In addition to that, they are on the $(\tilde{S}_{2,1}^E, \tilde{S}_{3,1}^E)$ plane because $\tilde{S}_{1,1}^E$ is constant. The points $\tilde{S}_{1,1}^E = (\pm 1, 0, 0)$ are the fixed points, which reflects that the RP and the AP modes are orthogonal modes. The DOP is a monotonic decreasing function of $\tilde{z}$. Its value at $\tilde{z} \to \infty$ is $|\tilde{S}_{1,1}|$, which is ascribed to the fact that the RP and the AP modes propagate independently with the different Rayleigh lengths.
5.3 Nonlinear propagation of axisymmetrically polarized pulses in a uniaxial crystal

5.3.1 Third-order nonlinear polarization

In order to include nonlinearity into the wave equation, we write the electric flux density $D$ as

$$D = \epsilon E + P^{NL}, \quad (5.3.1)$$

where $P^{NL}$ is the nonlinear polarization. We here consider the degenerate third-order nonlinear optical susceptibility $\chi^{(3)}(-\omega; \omega, \omega, -\omega)$ of 422, 4mm, 4/mmm, 42m, 432, m3m, 3m, -3m, 32, 622, 6mm, 6/mmm, -6m2 and isotropic crystals [158]. Thus, the transverse nonlinear polarization $P^{NL}_\perp = (P_x, P_y)$ can be written [98, 159] as

$$P_x = 2\epsilon_0 n_o n_2 E^2 \left( |E_x|^2 + \gamma_X |E_y|^2 \right) E_x + \gamma_F E_x^* E_y^2,$$

$$P_y = 2\epsilon_0 n_o n_2 E^2 \left( |E_x|^2 + |E_y|^2 \right) E_y + \gamma_F E_y^* E_x^2,$$  \quad (5.3.2, 5.3.3)

where $n_o$ is the ordinary refractive index, $n_2^E$ is the nonlinear refractive index, $\gamma_X$ is the ratio of the self-phase modulation (SPM) to the cross-phase modulation (XPM) and $\gamma_F$ is the ratio of the SPM to the four-wave mixing (FWM). They are defined as

$$n_o = \sqrt{\epsilon_0}, \quad (5.3.4)$$

$$n_2^E n_o \frac{8}{3} = \chi^{(3)}_{xxxx}, \quad (5.3.5)$$

$$\gamma_X n_2^E n_o \frac{8}{3} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} = 2\chi^{(3)}_{xxyy}, \quad (5.3.6)$$

$$\gamma_F n_2^E n_o \frac{8}{3} = \chi^{(3)}_{xyyx}, \quad (5.3.7)$$

where $\chi^{(3)}_{ijkl} \equiv \chi^{(3)}_{ijkl}(-\omega; \omega, \omega, -\omega)$ is the degenerate third-order nonlinear susceptibility tensor. In trigonal (3m, -3m, 32), hexagonal (622, 6mm, 6/mmm, -6m2) and isotropic crystals, the condition $\gamma_F = 1 - \gamma_X$ is added. Since we propagate a pulse beam along the optic axis of a uniaxial crystal, the axial components $E_z$ and $P_z$ are assumed to remain so small that they can be negligible [105].
5.3.2 Paraxial nonlinear wave equation

When the pulse has many cycles in the pulse duration $\tau$ ($\omega \tau \gg 1$), we can apply an approximation of the quasi-monochromatic light wave:

$$ \mathbf{E} = \tilde{\mathbf{E}} e^{i(kz - \omega t)}, \quad (5.3.8) $$

$$ P_{\perp}^\text{NL} = \tilde{P}_{\perp}^\text{NL} e^{i(kz - \omega t)}. \quad (5.3.9) $$

Thus we obtain the paraxial nonlinear wave equation [114, 154]:

$$ 2i k \sqrt{\epsilon_0} \partial_z \tilde{E}_\perp + \nabla_\perp^2 \tilde{E}_\perp - \alpha \nabla_\perp (\nabla_\perp \cdot \tilde{E}_\perp) + \frac{k^2}{\epsilon_0} \tilde{P}_{\perp}^\text{NL} = 0, \quad (5.3.10) $$

where $kw \gg 1$ and $|1 - \epsilon_o/\epsilon_e| \gg \epsilon_o n_o n_2|E_{x,y}^2|$ are assumed. By using the AxP representation, Eq. (5.3.10) is expressed as

$$ 2ik n_o \partial_z \tilde{E}_r = - \beta \left( \partial_r^2 + \frac{1}{r} \partial_r - \frac{1}{r^2} \right) \tilde{E}_r \right. $$

$$ \left. - 2k^2 n_0 n_2^E \left[ (|\tilde{E}_r^*|^2 + \gamma_X |\tilde{E}_\phi|^2) \tilde{E}_r + \gamma_F \tilde{E}_\phi^2 \tilde{E}_r^* \right] \right), \quad (5.3.11) $$

$$ 2ik n_o \partial_z \tilde{E}_\phi = - \left( \partial_r^2 + \frac{1}{r} \partial_r - \frac{1}{r^2} \right) \tilde{E}_\phi $$

$$ - 2k^2 n_0 n_2^E \left[ (\gamma_X |\tilde{E}_r|^2 + |\tilde{E}_\phi|^2) \tilde{E}_\phi + \gamma_F \tilde{E}_r^2 \tilde{E}_\phi^* \right] \right), \quad (5.3.12) $$

where $\beta = \epsilon_o/\epsilon_e = (n_o/n_e)^2$. In contrast to the paraxial linear wave equation (Eq. (5.2.14)), these equations are mutually dependent and have nonlinear terms as follows:

1. the SPM terms of $\tilde{E}_r$ or $\tilde{E}_\phi$,
2. the XPM terms between $\tilde{E}_r$ and $\tilde{E}_\phi$,
3. the FWM terms between $\tilde{E}_r$ and $\tilde{E}_\phi$.

The SPM and XPM effects are regarded as just the nonlinear phase modulation. In contrast to that, the FWM effect involves energy exchange between the RP and the AP modes, thus we expect the value change of $S_{1,1}^E$ while it never changes in the linear propagation regime.

5.4 Experimental

5.4.1 Setup

Figure 5.4.1 shows an experimental setup for the nonlinear propagation of AxP modes. The light source used was a Ti:Sa laser amplifier (center wave-
Figure 5.4.1: Experimental setup. Wedge, a wedge laser window; BPF, a bandpass filter (800±5 nm); ND1, a neutral density filter; M1-3, mirrors; L1-6: lens (L1, \(f = 100\) mm; L2, \(f = 50\) mm; L3, \(f = 60\) mm; L4, \(f = 35\) mm; L5, \(f = 200\) mm; L6, \(f = 80\) mm); PH, a pinhole with a 100 \(\mu\)m hole; PBS1,2, polarizing beam splitters; POL, a polarizer; HWP1,2, achromatic half-wave plates; QWP1,2, achromatic quarter-wave plates; SPP, a spiral phase plate \((l = -1)\); NC, a nonlinear crystal \((c\)-cut Calcite crystal); CCD, a charge-coupled-device camera.
length 800 nm, bandwidth of ∼40 nm, pulse duration ∼25 fs, and repetition rate 1 kHz). In order to attenuate the pulse energy, we let the pulse beam reflect at a wedge laser window (Wedge), and used the reflected pulse beam. The large part of the pulse energy transmits the wedge window and is dumped. The pulse beam is passed through and a bandpass filter (BPF, center wavelength 800 nm, bandwidth of 10 nm) and a neutral density filter (ND1), which made the pulse duration ∼120 fs and the pulse energy ∼1 µJ. A spatial filter composed of relay lens of L1 (f = 100 mm) and L2 (f = 50 mm), and a pinhole (PH, hole size was 100 µm) improved the beam quality. A pair of a half-wave plate (HWP1) and a polarizing beam splitter (PBS1) enabled us to adjust the pulse energy. A polarizer (POL), an achromatic quarter-wave plate (QWP1) and a spiral phase plate (SPP) converted the pulses into |s⟩=1 |l⟩=−1 OV pulses. The pulses were focused by a lens (L3), following which the focused pulses with the Rayleigh length of 0.5 mm propagated in a c-cut Calcite crystal (NC). We can consider that the beam divergence is small enough to use the paraxial approximation. The pulse beam axis was set to be parallel to the optic axis of NC. The position of NC on the z-axis (the beam axis) can be controlled by a motorized linear stage. A lens (L4) collimated the pulse beam. By using the Stokes polarimetry system [110] constructed by a pair of a quarter-wave plate (QWP2) or a half-wave plate (HWP2), a polarizing beam splitter (PBS2) and a charge-coupled-device camera (CCD), we obtained polarization distribution of the output pulse beam. Lenses of L5 and L6 form a relay lens system.

5.4.2 Results

We investigated the focus position (or the crystal position) dependence of the normalized ESPs and the DOP-SD. The focus position is characterized z_F, which is the focus position when the crystal is removed (Fig. 5.4.2). There are three characteristic focal positions; One is the position where the focus is at the input facet of the crystal: z_F = 0 (Fig. 5.4.2 (a)), another is the position where the focus of the AP mode is at the output facet of the crystal (Fig. 5.4.2 (b)):

\[ n_o z_F^A = 1.65 z_F^A = L, \]  

(5.4.1)

the other is the position where the focus of the RP mode is at the output facet of the crystal (Fig. 5.4.2 (c)):

\[ \frac{n_e^2}{n_o} z_F^R = 1.33 z_F^R = L. \]  

(5.4.2)
Figure 5.4.2: Trajectories of beam widths for the input beam (red line), the RP mode (green line) and the AP mode (yellow line). (a) The focus of input beam corresponds to the input facet of the crystals \( z_F = 0 \) mm. The focus of the AP mode corresponds to the output facet of (b) 2 mm-thick crystal \( z_F \sim 1.2 \) mm and (c) that of 5 mm-thick crystal \( z_F \sim 3.0 \) mm.

From the paraxial nonlinear wave equation (Eqs. (5.3.11) and (5.3.12)), the FWM effect can change the value of \( \tilde{S}_{E,1,1} \). In addition to that, nonlinear effects generally involve nonlinear focusing effects, which makes the change of the effective Rayleigh and the effective propagation length. Being a monotonically decreasing function (Eq. (5.2.24)) in the linear propagation, DOP-SD can be a good probe to sense the change of effective propagation length. Thus, we analyze the difference of \( \tilde{S}_{E,1,1}^{(P)} \) and \( P_1^{(\text{space})} \) from the values of them in the linear propagation \( \tilde{S}_{E,1,1}^{(P),L} \) and \( P_1^{(\text{space}),L} \), respectively:

\[
\Delta \tilde{S}_{E,1,1}^{(P)}(z_F) = \tilde{S}_{E,1,1}^{(P)}(z_F) - \tilde{S}_{E,1,1}^{(P),L}, \quad (5.4.3)
\]

\[
\Delta P_1^{(\text{space})}(z_F) = P_1^{(\text{space})}(z_F) - P_1^{(\text{space}),L}. \quad (5.4.4)
\]

The experimental results are shown in Fig. 5.4.3. The experiment was conducted under the two conditions of the pulse energy: 0.81 µJ and 0.39 µJ, and the two crystal thickness: 2 mm and 5 mm. From Fig. 5.4.3 (a) and (b), the energy ratio between the RP and the AP changed in the region of \( 0 \lesssim z_F \lesssim z_F^A \). Moreover, the value of \( \Delta \tilde{S}_{E,1,1}^{(P)}(z_F) \) was wholly proportional to the pulse energy and has a positive peak. In Fig. 5.4.3 (c) and (d), the value of \( \Delta P_1^{(\text{space})}(z_F) \) also has the dependence of pulse energy but has not
only the positive peak at $z_F = 0$ but also the negative peak at $z_F = z_F^A$.

Interpretation of the experimental results is made with difficulty just by looking at polarization distributions such as Fig. 5.4.3 (e)-(h). In good contrast, the ESPs and the DOP-SD make the experimental results easy to understand.
Figure 5.4.3: Experimental results for nonlinear propagation in uniaxial crystals (c-cut Calcite) of 2 mm thickness ((a) and (c)) and 5 mm thickness ((b) and (d)) [114]. The cyan bars represent the focus position where the input beam is at the input facet ($z_F = 0$). The orange and green bars correspond to the focus positions $z_F$ where the foci of the AP and the RP modes are at the output facet, respectively. (e)-(h) are polarization distributions at $z_F = -2.0$ mm (e), 0.0 mm (f), 1.2 mm (g), 3.0 mm (h) when the crystal thickness was 5 mm and the pulse energy was 0.81 $\mu$J. All the polarization distributions are colored under the rule in Fig. 4.3.3.
5.5 Simulation

We carry out numerical simulations to analyze experimental results. We choose the Runge-Kutta and pseudo-spectrum method for $z$- and $r$-differentions, respectively. Since wave equations (Eqs. (5.3.11) and (5.3.12)) have off-diagonal components, the split-step Fourier method cannot be used here [105]. With $\tilde{z} = z/z_0$ and $\tilde{r} = r/w_0$, we normalize the wave equations as follows:

$$\partial_{\tilde{z}} \tilde{E}_r = i \left\{ \frac{n_o}{4n_e^2} \left( \frac{\partial^2}{\tilde{r}^2} + \frac{1}{\tilde{r}} \frac{\partial}{\tilde{r}} - \frac{1}{\tilde{r}^2} \right) \tilde{E}_r + k z_0 n_e E^2 \left[ (|\tilde{E}_r|^2 + \gamma_X |\tilde{E}_\phi|^2) \tilde{E}_r + (1 - \gamma_X) \tilde{E}_\phi^2 \tilde{E}_r^* \right] \right\}, \tag{5.5.1}$$

$$\partial_{\tilde{z}} \tilde{E}_\phi = i \left\{ \frac{1}{4n_o} \left( \frac{\partial^2}{\tilde{r}^2} + \frac{1}{\tilde{r}} \frac{\partial}{\tilde{r}} - \frac{1}{\tilde{r}^2} \right) \tilde{E}_\phi + k z_0 n_o E^2 \left[ (\gamma_X |\tilde{E}_r|^2 + |\tilde{E}_\phi|^2) \tilde{E}_\phi + (1 - \gamma_X) \tilde{E}_r^2 \tilde{E}_\phi^* \right] \right\}, \tag{5.5.2}$$

where $\gamma_F = 1 - \gamma_X$ is applied because calcite crystals (3m) are used in the experiment.

We used a spiral phase plate to generate $|s=+1\rangle |l=1\rangle$ OV pulses from $|s=+1\rangle |l=0\rangle$ Gaussian pulses in the experiment. The generated beam $\tilde{E}^{(gen)}(e_r^1 + i e_\phi^1)$ is referred to as a point vortex [160], a hypergeometric-Gaussian mode [161] or a Kummer beam [162] described as

$$\tilde{E}^{(gen)} = A^{(gen)} \sum_{p=0}^{\infty} \frac{\pi(2p-1)!!}{2\sqrt{2(p+1)p!}(2p)!!} u_{LP}^{1p}.$$  

We perform simulations in two cases; one is the case where the input transverse electric field $\tilde{E}_L(r, \phi, z = 0)$ is described by Eq. (5.2.19) (the fundamental mode in $r$-direction in Eq. (5.5.3)), and the other is the case where $\tilde{E}_L(r, \phi, z \leq 0)$ is expressed by the mixed mode with $p = 0, 1$ and 2 in Eq. (5.5.3). We assume that the absolute value of the fundamental mode is normalized so that the maximum of the absolute value of the input beam at its focus is unity ($A_r = -i A_\phi = |\exp(1)/2|^{1/2}$ in Eq. (5.5.3)), and the total intensity of the mixed mode in the beam cross section is the same as that of the fundamental mode ($A^{(gen)} = 8\sqrt{2}/(5\sqrt{3\pi})$).

Calculations were done under the condition that $k z_0 n_e^2 = 0.1$, $\gamma_X = 2/3$, the wavelength of OV is 800 nm, and a calcite ($n_o = 1.64, n_e = 1.48$ [163]) is used as a nonlinear crystal. Figure 5.5.1 (a), (c) and (b), (d) show the three-dimensional plots of $\Delta S^E_{1,1}(z_F/z_0)$ and $\Delta D_{1}^{(space)}(z_F/z_0)$, respectively.

From Fig. 5.5.1 (a) and (c), the energy transfer between the AP and the RP modes occurs at the propagation length in the vicinity of their foci and
Figure 5.5.1: Simulation results for $kz_0n_2^E = 0.1, \gamma_X = 2/3$. Focus position dependence of $\Delta S^E_{1,1}(z)$ and $\Delta V^E_{\text{space}}(z)$. (a), (b) the fundamental mode ($p = 0$) input and (c), (d) the mixed-mode ($p = 0, 1$ and 2) input. The orange and green dashed lines represent focus position of the AP and the RP modes, respectively.
Figure 5.5.2: Simulation results for focus position dependence of $\Delta S_{1,1}^E$ and $\Delta P_{1}^{\text{space}}$ at two standardized crystal lengths. (a) and (c) $z = 4z_0$; (b) and (d) $z = 10z_0$ $(\gamma_X = 2/3)$. These results are for the mixed-mode ($p = 0, 1$ and $2$) input. The cyan bars represent the focus position where the input beam is at the input facet ($z_F = 0$). The orange and green bars correspond to the focus positions $z_F$ where the foci of the AP and the RP modes are at the output facet, respectively.
the value of $\Delta S_{1,1}^{E}$ are conserved after the transfer ($z_F \gtrsim z_A^{\text{out}}$) with the propagation length. The width of the $\Delta S_{1}^{E}$ peak in the mixed-mode beam (Fig. 5.5.1 (a)) is apparently narrower than that in the fundamental mode beam (Fig. 5.5.1 (c)). $\Delta P_1^{(\text{space})}$ has a positive peak around $z_F = 0$ and a negative peak on orange dashed lines ($z = (c_\varepsilon^2/\varepsilon_0)^{1/2} z_R^{\text{out}}$) in Fig. 5.5.1 (b) and (d).

In Fig. 5.5.2, we draw the cross-sectional view of Fig. 5.5.1 as a function of $z_F$ at propagation distances of $z = 4z_0$ ($\sim 2\text{mm}$) and $z = 10z_0$ ($\sim 5\text{mm}$). The experimental results (Fig. 5.4.3) agree with the simulation results using the mixed mode rather than those using the fundamental mode. The differences of the peak values $\Delta S_{1}^{E}$ at $z = 4z_0$ and at $z = 10z_0$ in the mixed-mode case are small, resembling the experimental results.

5.6 Discussion

First, we discuss nonlinear $\tilde{S}_{1,1}^{E}(z_F)$ change in the experimental result. The value of $\Delta S_{1}^{E}(z_F)$ changes in the region of $0 \lesssim z_F \lesssim z_A$. This fact means that there was energy conversion between the $\tilde{E}_r$ and the $\tilde{E}_\phi$ components in the crystal when the foci of the RP and the AP modes are in the crystal. In taking the change is proportional to the pulse energy into account, this phenomenon apparently reflects the FWM effect between the $\tilde{E}_r$ and the $\tilde{E}_\phi$ components, which agrees with the simulation results. This nonlinear energy conversion occurred from the $\tilde{E}_\phi$ component to the $\tilde{E}_r$ component in this experimental condition. Since Eqs. (5.3.11) and (5.3.12) are invariance under a permutation such that

$$n_o \rightarrow n_e^2/n_o,$$ 
$$\tilde{E}_r \leftrightarrow \tilde{E}_\phi,$$ 

the direction of nonlinear energy conversion is determined by the sign of the anisotropy of the crystal. Hence, we can control or switch the transfer direction by using a positive uniaxial crystal instead of a negative crystal.

It should be noted that a phase mismatch due to Gouy phase shift $\Psi_G(z)$ affects the $\Delta S_{1,1}^{E}$ behavior. From Eq. (5.3.11), the relative Gouy phase shift between the RP component $\tilde{E}_r$ and the RP FWM polarization $\tilde{E}_\phi^2 \tilde{E}_r^*$ is $\Psi_G - (2\Psi_\phi^r - \Psi_G^r) = 2(\Psi_G^r - \Psi_G^\phi)$, where $\Psi_G^r$ and $\Psi_G^\phi$ are Gouy phase shift for the RP and the AP components, respectively. If we assume that the transverse electric field with the RP and the AP components is roughly described by Eq. (5.2.20), the relative Gouy phase difference between two components is $4(\arctan((z/n_o - z_F)/z_0) - \arctan((zn_o/n_e^2 - z_F)/z_0))$. Figure 5.6.1 depicts the
Figure 5.6.1: Relative Gouy phase difference between $\tilde{E}_r$ and $\tilde{E}_\phi^2 \tilde{E}_r^*$. We assume that $\tilde{E}_r$ and $\tilde{E}_\phi$ are described by Eq. (5.2.20). The orange and green dashed lines represent focus position of the AP mode and the RP mode, respectively.

relative Gouy phase difference as a function of the focal length position $z_F$ and the normalized propagation distance. The relative Gouy phase difference varies in the vicinity of the two foci of the AP and the RP pulses. It flips the energy flow direction between the foci of the AP and RP modes if the distance of these foci is over $\sim z_0$. From the simulation result of the fundamental mode (Fig. 5.5.1 (a)), the region where the flip occur corresponds to that of $6.5z_0 \lesssim z_F$, where $\Delta S_{1,1}^E(z_F/z_0)$ decreases between the foci of the AP and RP modes. In the mixed mode, the region where the flip occur spreads ($3.5z_0 \lesssim z_F$). These findings explain the calculation results that the width or distribution of the positive peak is narrower in the mixed-mode case than that in the fundamental case, as follows. In the experiment, the pulse contains all radial-index $p$-LG mode with an azimuthal index of $l = -1$. The higher $p$-LG mode undergoes the larger Gouy phase shift (Eq. (2.1.17)), which complicates the energy flow direction. As we can see the numerical results in Fig. 5.5.1 (a) and (c), the width or distribution of the $\Delta S_{1,1}^E(z_F/z_0)$ indeed changes with the contamination of just a few $p \neq 0$ modes. It indicates that nonlinear propagation effect of optical vortex is affected by the mixture of higher $p$ modes and that the Gouy phase shifts with mode mixture modify the energy transfer between the RP and AP modes.

The nonlinear DOP-SD change can be basically understood from the change of the effective propagation length. Nonlinear effects, in particular the SPM and the XPM effects, cause nonlinear focusing in calcite crystals, which can make a beam divergence wider or narrower than that in the linear propagation. The beam divergence change depends on the position of $z_F$. 

64
Figure 5.6.2: Divergence change made by nonlinear effects decreases (Case 1; $z_F^{A,R} \lesssim 0$) or increases (Case 2; $0 \lesssim z_F^{A,R} \lesssim L$) the effective normalized propagation length, which causes the positive or negative change of DOP-SD $P_1^{(\text{space})}$.

(Fig. 5.6.2). When $z_F^{A,R} \lesssim 0$, the nonlinear focusing effect makes beam divergence narrow. In contrast to that, the divergence expands in the region of $0 < z_F^{A,R} \lesssim L$. The divergence change can be regarded as the change of the effective Rayleigh length. In the linear propagation, the DOP-SD $P_1^{(\text{space})}$ depends on the Rayleigh length $z_0$ when the crystal length $L$ (or $z$) is fixed (Eq. (5.2.24)). Since the DOP-SD $P_1^{(\text{space})}$ is a monotonically decrease function of $z/z_0$, the positive and the negative peaks of $\Delta P_1^{(\text{space})}$ can be ascribe to the nonlinear focusing in the crystal; the sign of $\Delta P_1^{(\text{space})}$ is positive or negative when the focus is around the input facet $z_F^{A,R} \lesssim 0$ (Fig. 5.6.2, Case 1) or in the crystal $0 < z_F^{A,R} \lesssim L$ (Fig. 5.6.2, Case 2), respectively.
Chapter 6
Summary and prospect

In this thesis, the framework to measure CP states full-quantitatively has been established through newly introducing of the ESPs, their DOP and the DOP-SD. Moreover, the experimental demonstrations of utilizing the ESPs and the DOP-SD were conducted. We summarize this thesis and give some prospects of utilizing the ESPs and the DOP-SD.

**ESPs, DOP-SD and PBP on EPS**  We have newly introduced the ESPs and their DOP or DOP-SD in order to evaluate CP states full-quantitatively. Unlike the HOSPs, the ESPs are capable for the definition of the DOP, which complies the traditional framework of Stokes parameters. To the best of our knowledge, the ESPs and their DOP (DOP-SD) are the unique parameters which enable us to make full-quantitative measurements of CP beams.

Moreover, we introduced the EPS of the ESPs and discussed the PBP for the EPS. We obtained the general representations of the “Hamiltonians” of the Maxwell-Schrödinger equations for arbitrary precession motions on the EPS. After that, we consider the PBP made by integer order $q$-plates and conclude that the PBP is subject to the same mathematics as the PBP in the spin-1/2 Hamiltonian system. In order to discuss the PBP for the half-integer order $q$-plates, we need to furthermore extend the order $l$ of the ESPs to half-integer, which is a future work of this topic. In addition to that, it is an interesting research theme to explore the PBP when the laser beam passes through a sequence of various $q$-plates with different order $q$.

**Full-quantitative measurement of CP pulses**  We have experimentally demonstrated full-quantitative measurement with the ESPs and DOP-SD through the characterization of CP broadband pulses (Chap. 4) and the analysis for the nonlinear propagation of CP pulses in a uniaxial crystal (Chap. 5). The experimental acquirement of polarization distributions are
done with a simple optical system composed of a half-wave plate, a quarter-wave plate, a polarizer and a CCD camera.

In Chap. 4, we have shown that the arbitrary CP broadband pulse states with high symmetry and low spectral dependence were generated from the system employing the coherent beam combining system. Especially, the good agreement between the experimental and simulation results indicates that the degradation in DOP-SD is ascribed to the deformation of incident OV pulses and the contamination of perpendicular polarized components at the polarizing optical components, and ensures that we can quantitatively investigate even the small differences of rotational symmetry of polarization distributions or the small contamination of unwanted modes by using DOP-SD. At least $\Delta P_{1,2}^{\text{space}} \simeq 0.02$ is significant and detectable in our measurement system. Though the earlier studies have not taken account of DOP-SD, DOP-SD as well as ESPs is an important parameter for full-quantitative characterization of CP states.

It has been pointed out that precise measurement of polarization state is important in quantum information [164]. Applications using polarized pulses such as material processing [165], magneto-optical storage [138] and nonlinear spectroscopic polarimetry [51] also need to know their polarization states precisely. Using CP pulses instead of the conventional uniform polarized pulses is a manner to extend the degree of freedom in these applications, which have been already demonstrated in quantum information science [40, 41, 43], material processing [18] and nonlinear spectroscopic polarimetry [51]. Our fully-quantitative measurement method for CP pulses hence can improve the sophistication of these applications.

In Chap. 5, the nonlinear effects occurring in a uniaxial crystal was discussed by using the ESPs and the DOP-SD. The value changes of the ESPs and the DOP-SD from the values in the linear propagation respectively reflect the nonlinear energy conversion phenomenon and the nonlinear focusing effects. Although our experimental set up is similar to $z$-scan methods and D4$\sigma$ methods [166–170] for determining the value of nonlinear susceptibility, our experiment differs from them in using the AxP pulses and taking notice of the changes of CP states through the ESPs and the DOP-SD. Thus, we suggest a new method to investigate nonlinear effects in crystals.

In the present thesis, we have considered only the DOP-SD with single order of $l$. As future work on the exploring linear and nonlinear interaction between matter and laser beams, measuring the DOP-SD spectrum of the output beam may be a powerful tool for analyzing spatial rotational symmetry of the material. For example, we can apply the analysis of the DOP-SD spectrum to the spin-to-orbital angular momentum conversion trigged by scattering in a small dipole particle [171–174].
Appendix A

Polarization of light

In this chapter, we define polarization, obtain the form of Jones matrices for some important optic elements, and introduce the method of rotating-retarder type imaging polarimetry.

A.1 Definition of polarization

We discuss polarization of a monochromatic electromagnetic light wave whose electric components are described by

\[
E_\perp = \begin{pmatrix} E_x \\ E_y \end{pmatrix} = Ae^{i(kz-\omega kt)} \begin{pmatrix} \cos \alpha \\ \sin \alpha e^{i\delta} \end{pmatrix},
\]

(A.1.1)

where \( A \) is the complex amplitude, and \( \alpha \) and \( \delta \) are the angles to characterize the state of the electromagnetic field (\( 0 \leq \alpha < \pi, -\pi/2 \leq \delta \leq \pi/2 \)). The trajectory of the real electric vector \( \Re\{E_\perp\} \) satisfies [110, 175]

\[
\left( \frac{\Re[E_x]}{|A| \cos \alpha} \right)^2 + \left( \frac{\Re[E_y]}{|A| \sin \alpha} \right)^2 - 2 \frac{\Re[E_x]}{|A| \cos \alpha} \frac{\Re[E_y]}{|A| \sin \alpha} \cos \delta = \sin^2 \delta.
\]

(A.1.2)

When \( \delta = 0 \), this equation is transformed into

\[
\Re[E_y] = \tan \alpha \cdot \Re[E_x],
\]

(A.1.3)

which gives a linear trajectory in the \( (\Re[E_x], \Re[E_y]) \) plane. We call this polarization state a linear polarized state. When \( \delta = \pm \pi/2 \) and \( \alpha = \pi/4 \), Eq. (A.1.2) describes the equation of circle:

\[
\Re[E_x]^2 + \Re[E_y]^2 = |A|^2.
\]

(A.1.4)
Figure A.1.1: Elliptically polarized state.

We call this polarization state a circularly polarized state. In order to discuss the general trajectory when $\delta \neq 0$, we introduce the $\left( \Re\left[E_\xi\right], \Re\left[E_\eta\right] \right)$ plane described by

$$\left( \Re\left[E_\xi\right], \Re\left[E_\eta\right] \right) = R_{-\alpha} \left( \Re\left[E_x\right], \Re\left[E_y\right] \right).$$

(A.1.5)

Hence Eq. (A.1.2) is converted into

$$\Re\left[E_\xi\right]^2 + \frac{\Re\left[R_\eta\right]^2}{(1-\cos\delta)^2} = |A|^2 \frac{1 + \cos\delta}{2},$$

(A.1.6)

which gives the equation of ellipse (Fig. A.1.1). This state is named an elliptically polarized state.

When the polarization state is a circularly or elliptically polarized state, the direction of the rotation is respectively clockwise ($-\pi/2 \leq \delta < 0$) or counterclockwise ($0 < \delta \leq \pi/2$) because the sign of the velocity vector at
\( E_\perp (t = 0, z = 0) = |A| \left( \cos \alpha, \sin \alpha \cos \delta \right) \) is determined by \( \delta \):

\[
\partial_t \Re [E_\perp] (z = 0)|_{t=0} = |A| \omega \begin{pmatrix} 0 \\ \sin \alpha \sin \delta \end{pmatrix}.
\] (A.1.7)

We respectively define the clockwise and counterclockwise direction from \(+z\) axis as the right and left directions.

Generally, polarization of light wave can be described by a Jones vector \( J \) as follows:

\[
J = \begin{pmatrix} \sin \alpha \\ \cos \alpha e^{i\delta} \end{pmatrix}.
\] (A.1.8)

### A.2 Jones matrix

Jones matrices express transformations of Jones vectors made by optic elements [110, 159]. We show the form of a Jones matrix of a retarder where the fast axis (\( \xi \) axis) and the slow axis (\( \eta \) axis) are defined by

\[
\begin{pmatrix} \xi \\ \eta \end{pmatrix} = R_{-\theta} \begin{pmatrix} x \\ y \end{pmatrix}.
\] (A.2.1)

The Jones matrix is written as

\[
\hat{O}_{\text{ret}} (\delta, \theta) = R_\theta \begin{pmatrix} e^{-i\delta/2} & 0 \\ 0 & e^{i\delta/2} \end{pmatrix} R_{-\theta}
= \begin{pmatrix} \cos \frac{\delta}{2} - i \cos 2\theta \sin \frac{\delta}{2} & -i \sin 2\theta \sin \frac{\delta}{2} \\ -i \sin 2\theta \sin \frac{\delta}{2} & \cos \frac{\delta}{2} + i \cos 2\theta \sin \frac{\delta}{2} \end{pmatrix}
= \cos \frac{\delta}{2} \sigma_0 - i \sin 2\theta \sin \frac{\delta}{2} \sigma_1 - i \cos 2\theta \sin \frac{\delta}{2} \sigma_3,
\] (A.2.2)

where \( \delta \) is the retardance.

The Jones matrices for a quarter-wave plate (\( \delta = \pi/4 \)) and a half-wave plate (\( \delta = \pi/2 \)) are respectively described as

\[
\hat{O}_{\text{QWP}} (\theta) = \hat{O}_{\text{ret}} (\pi/2, \theta) = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} - i \begin{pmatrix} \cos 2\theta & \sin 2\theta \\ \sin 2\theta & -\cos 2\theta \end{pmatrix}
= \frac{1}{\sqrt{2}} (\sigma_0 - i \sin 2\theta \sigma_1 - i \cos 2\theta \sigma_3),
\] (A.2.5)

\[
\hat{O}_{\text{HWP}} (\theta) = \hat{O}_{\text{ret}} (\pi, \theta) = -i \begin{pmatrix} \cos 2\theta & \sin 2\theta \\ \sin 2\theta & -\cos 2\theta \end{pmatrix}
= -i \sin 2\theta \sigma_1 - i \cos 2\theta \sigma_3.
\] (A.2.6)
A.3 Rotating-retarder type imaging polarimetry

The rotating-retarder type imaging polarimeter [110, 176], which is a system to obtain polarization distributions of laser beams, is composed of a retarder, a polarizer and a CCD camera (Fig. A.3.1). We define $\theta$ as the angle of the fast axis of the retarder from $x$ axis, and $d$ is the thick of the retarder. We consider the transverse electric field before the retarder ($z < 0$) is described as

$$E_{\perp}^{\text{in}} = E_{\perp}^{\text{in}}(r, t) = \begin{pmatrix} E_{x}^{\text{in}}(r, t) \\ E_{y}^{\text{in}}(r, t) \end{pmatrix} \quad (z < 0), \quad \quad (A.3.1)$$

where the retarder is allocated from $z = 0$ to $z = d$. The transverse electric field after the retarder is expressed as

$$E_{\perp}^{\text{out}} = E_{\perp}^{\text{out}}(r, t) = \begin{pmatrix} E_{x}^{\text{out}}(r, t) \\ E_{y}^{\text{out}}(r, t) \end{pmatrix} = \hat{O}_{\text{ret}}(\delta, \theta)E_{\perp}^{\text{in}} \quad (z > d). \quad \quad (A.3.2)$$

The coherency matrix of the transverse electric field after the retarder is calculated to be

$$(E_{\perp}^{\text{out}} E_{\perp}^{\text{out}\dagger}_t) = \begin{pmatrix} \langle |E_{x}^{\text{out}}|^2 \rangle_t & \langle E_{x}^{\text{out}} E_{y}^{\text{out}\dagger} \rangle_t \\ \langle E_{y}^{\text{out}\dagger} E_{x}^{\text{out}} \rangle_t & \langle |E_{y}^{\text{out}}|^2 \rangle_t \end{pmatrix} = \langle \hat{O}_{\text{ret}} E_{\perp}^{\text{in}} E_{\perp}^{\text{in}\dagger} \hat{O}_{\text{ret}} \rangle_t = \hat{O}_{\text{ret}}(s(x, y) \cdot \sigma^{xy})\hat{O}_{\text{ret}}^{\dagger} = s_0 \sigma_0 \hat{I}$$
\[ + \left[ \sin 4\theta \sin^2 \frac{\delta}{2} s_1 + \left( \cos^2 \frac{\delta}{2} - \cos 4\theta \sin^2 \frac{\delta}{2} \right) s_2 - \cos 2\theta \sin \delta s_3 \right] \sigma_1 \\
+ \left[ - \sin 2\theta \sin \delta s_1 + \cos 2\theta \sin \delta s_2 + \cos \delta s_3 \right] \sigma_2 \\
+ \left[ \left( \cos^2 \frac{\delta}{2} + \cos 4\theta \sin^2 \frac{\delta}{2} \right) s_1 + \sin 4\theta \sin^2 \frac{\delta}{2} s_2 + \sin 2\theta \sin \delta s_3 \right] \sigma_3, \]

(A.3.3)

where \( s(x, y) \) is a Stokes vector of the transverse electric field before the retarder. Thus, the intensity distribution captured by the CCD camera is proportional to

\[
\langle |E_{x}^{\text{out}}(r, t)|^2 \rangle_t = \frac{1}{2} \left[ s_0 + \left( \cos^2 \frac{\delta}{2} + \cos 4\theta \sin^2 \frac{\delta}{2} \right) s_1 + \sin 4\theta \sin^2 \frac{\delta}{2} s_2 + \sin 2\theta \sin \delta s_3 \right],
\]

(A.3.4)

or

\[
\langle |E_{y}^{\text{out}}(r, t)|^2 \rangle_t = \frac{1}{2} \left[ s_0 - \left( \cos^2 \frac{\delta}{2} + \cos 4\theta \sin^2 \frac{\delta}{2} \right) s_1 - \sin 4\theta \sin^2 \frac{\delta}{2} s_2 - \sin 2\theta \sin \delta s_3 \right],
\]

(A.3.5)

when the polarizer is parallel to the \( x \) axis or the \( y \) axis, respectively.

When we obtain at least four intensity distributions of different \( \theta \) satisfying linear independent of Eqs. (A.3.4) or (A.3.5), we can reconstruct the distribution of the Stokes parameters \( s(x, y) \).
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Glossary

4-f SLM  spatial light modulator in the 4-f configuration.

AP  azimuthally polarized.
AxP  axisymmetrically polarized.
BEC  Bose-Einstein condensation.
CCD  charge coupled device.
CP  cylindrically polarized.
CPLG  cylindrically polarized Laguerre-Gaussian.
CPS  conventional Poincaré sphere.
CSPs  conventional Stokes parameters.
CW  continuous wave.
DOP  degree of polarization.
DOP-SD  degree of polarization defined for the spatial distribution.
EPS  extended Poincaré sphere.
ESPs  extended Stokes parameters.
ESV  extended Stokes vector.
FWM  four-wave mixing.
HOSPs  higher-order Stokes parameters.
HSPs  hybrid Stokes parameters.
LCP  left-circularly polarized.
LG   Laguerre-Gaussian.
LP   linear polarized.

OAM orbital angular momentum.
OV optical vortex.

PBP Pancharatnam-Berry phase.
RCP  right-circularly polarized.
RP   radially polarized.

SAM spin angular momentum.
SLM  spatial light modulator.
SPM  self-phase modulation.

TAM total angular momentum.
Ti:Sa Ti:Sapphire.

TPPSU temporally-perfect-polarized but spatially unpolarized.
TSPP temporally- and spatially-perfect-polarized.
TU temporally unpolarized.

XPM cross-phase modulation.
Bibliography


