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Direct Generation of Vortex Laser Beams and Their Non-Linear Wavelength Conversion

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Abstract

Vortex laser beams are a technology that has revolutionised applications in micro- and nano-manipulation, micro-fabrication and super-resolution microscopy, and is now heralding advances in quantum communication. In order to service these, and emergent applications, the ability to generate powerful vortex laser beams with user-controlled spatial and wavefront properties, and importantly wavelength, is required. In this chapter, we discuss methods of generating vortex laser beams using both external beam conversion methods, and directly from a laser resonator. We then examine the wavelength conversion of vortex laser beams through non-linear processes of stimulated Raman scattering (SRS), sum-frequency generation (SFG), second harmonic generation (SHG) and optical parametric oscillation. We reveal that under different types of non-linear wavelength conversion, the spatial and wavefront properties of the vortex modes change, and in some cases, the spatial profile also evolve under propagation. We present a theoretical model which explains these dynamics, through decomposition of the vortex mode into constituent Hermite-Gaussian modes of the laser resonator.

Keywords: vortex laser, Laguerre-Gaussian modes, optical non-linear conversion, stimulated Raman scattering, sum-frequency generation, second harmonic generation, optical parametric oscillation, topological charge

1. Introduction

Vortex laser beams are characterised as having an annular spatial profile (with a central dark spot), and a wavefront which spirals along the direction of propagation of the beam (like a corkscrew). This is in contrast to a conventional Gaussian beam that has a plane wavefront [1–4]. The spatial, phase and wavefront properties of a Gaussian beam and vortex laser beams are shown in **Figure 1**.

Figure 1. Comparison of Gaussian and vortex laser beams showing spatial and wavefront profiles (Image modified, credit: E-karimi, Creative Commons Attribution-Share Alike 3.0 unported).

The topological charge (denoted l) of a vortex beam is equal to the integer number of a 2π phase change of the wavefront on any closed path around the propagation axis. Equivalently, one round trip around the phase surface leads to the next (or prior) coil with a pitch of $l\lambda$, where λ is the wavelength of the vortex beam [4]. Due to this spiral wavefront, vortex laser beams inherently carry orbital angular momentum (OAM), with each photon carrying orbital angular momentum of $\ell\hbar$ (where $\hbar$ is the reduced Planck constant). The magnitude of a vortex beam topological charge also impacts the size of the central dark spot relative to the overall size of the annular beam, with higher topological charge resulting in a larger spot. The direction of the wavefront, spiralling clockwise or anti-clockwise along the axis of propagation also indicates the sign of the topological charge, with positive (+) topological charge having a wavefront which spirals clockwise, and a negative (−) topological charge having a wavefront spiralling anti-clockwise [4].

It is not surprising that these unique characteristics of vortex laser beams have resulted in their application in a diverse range of applications. Indeed, vortex beams have revolutionised applications ranging from optical tweezing, in which micro- to nano-scale objects can be trapped and manipulated [3, 5], to fabrication of chiral nano-structures [6, 7], through to quantum communication [8]. Perhaps one of the most significant application of optical vortex beams has been in super resolution microscopy based on stimulated emission depletion (STED), a microscopy technique which has gained recognition with the Nobel Prize in Chemistry (2014). In STED microscopy, the annular profile of a vortex laser beam is used to selectively deplete the outer region of a fluorescing particle being imaged. This effectively

allows the resolution of fluorescence microscopy to exceed the diffraction limit [9, 10]. This host of applications therefore not only exploit the orbital angular momentum properties of the vortex beam, but also its annular spatial profile.

With the diversity of applications for vortex laser beams, comes the requirement for beams of the appropriate power and beam quality, topological charge, and importantly wavelength. As with 'standard' laser beams, there is continual drive to develop sources which can offer a diverse range of laser powers, beam quality and wavelengths. In this chapter, we detail methods of generating vortex laser beams, with a focus on methods of directly generating vortex beams from solid-state lasers (some of the most robust laser architectures currently available); and methods of wavelength-tuning vortex laser beams using non-linear optical processes. Here, we outline the dynamics of the processes by which these vortex beams can be manipulated, and the diversity of outputs that can be accessed.

2. Generation of vortex laser beams

There are a host of methods of generating vortex laser beams, however they can be broadly classified into two groups, those acting as convertors— taking a Gaussian beam laser beam and converting it into a vortex beam; and the direct generation methods whereby a vortex beam is produced directly from a laser resonator. The most commonly applied methods are those of the former group involving conversion of a Gaussian laser beam into a vortex beam.

2.1. Mode convertors

Conversion of a Gaussian laser beam into a vortex laser beam involves transformation of a plane wavefront into a spiral wavefront. This can be achieved using a number of methods including:

Phase plates: These are specially fabricated plates where the region through which the Gaussian beam propagates gradually increases in thickness (by stepping) in a spiral fashion. This transforms the plane wavefront of the Gaussian beam into a spiral wavefront by introducing a phase shift [11]. Through appropriate design of the phase plate, it is possible to use it to generate vortex laser beams with a desired topological charge.

Holograms/spatial light modulators (SLM): Much like phase plates, these elements act on the phase of the incident light to generate a spiral wavefront. Spatial light modulators also enable control over the order and sign of topological charge of the beam as they can be readily programmed [12, 13].

Cylindrical lens pairs: These act on higher-order Gaussian beams, typically Hermite-Gaussian beams. The cylindrical lens pair converts Hermite-Gaussian beams of a particular order, to their Laguerre-Gaussian counterparts [14]. As will be examined further in this chapter, Laguerre-Gaussian beams of the form LG_{0l} are vortex beams with topological charge l .

These methods are effective in converting Gaussian laser beams into vortex laser beams, however they are additional components which must be aligned precisely in addition to the actual

laser source itself. Phase plates and SLMs, in particular, suffer from relatively low conversion efficiency, and can be easily damaged at high laser powers. This limits their capacity to generate vortex laser beams with high power and high quality.

The ability to also generate vortex laser beams of a desired wavelength using these conversion methods is predicated on the initial Gaussian laser beam undergoing conversion, already being of the desired wavelength. In the following section, we detail methods of directly generating vortex laser beams from all solid-state laser systems, and demonstrate that these systems are capable of producing very high-power beams with excellent beam quality, without the need for additional, bulky and easily damaged beam converting components.

2.2. Direct generation methods

Methods of generating vortex laser beam output directly from a laser cavity rely on suppression of the lowest order Gaussian mode (TEM_{00}) and preferential oscillation of Laguerre-Gaussian modes of the form $LG_{0\ell}$. These Laguerre-Gaussian modes are vortex modes with topological charge (ℓ) [15, 16]. Suppression of the lowest order Gaussian mode can be achieved in a number of ways, including pumping the laser gain medium with an annular pump spot [17, 18], using intra-cavity defects [19–23] and thermal lensing [24, 25].

In this chapter, we focus on the generation of vortex laser beams using two of these methods, using an intra-cavity defect spot and thermal lensing. Intra-cavity defect spots are an effective means of very simply suppressing oscillation of the lowest-order Gaussian mode, to promote oscillation of an LG_{01} mode, and generate vortex laser output from an end-pumped solid-state laser. In these systems, the intra-cavity field is very intense, and by making use of this, we can readily investigate non-linear conversion of vortex beams. The use of thermal lensing to produce a vortex laser beam is also investigated in the context of a side-pumped slab laser. As will be detailed, this configuration is a very effective way of directly generating very high-power vortex laser beams, something which cannot be readily achieved using extra-cavity conversion components owing to component damage.

2.2.1. Intra-cavity defect spot

The use of an intra-cavity defect to suppress the lowest order oscillating mode (generally TEM_{00}) within a laser cavity has been achieved using a number of different laser systems, including He-Ne lasers [20] and solid-state lasers [19, 20–23]. The common feature of each of these systems is that an intra-cavity defect is used; such as engineered damage spots on resonator mirrors. Key to the effectiveness of this method is the ability for the defect to force oscillation of LG modes of the form $LG_{0\ell}$. The defect spot in effect breaks the symmetry of the laser resonator to enable oscillation of these LG modes. This requires accurate mode matching between the lowest order Gaussian mode (TEM_{00}) and the defect spot. As reported in the literature, the defect spot to cavity mode ratio is generally within the range ~ 0.15 – 0.2 for solid-state lasers [19–23]. The operating principle of this technique is shown schematically in **Figure 2**.

Here, we investigate generation of vortex laser emission from a continuous-wave, diode-end-pumped Nd:GdVO₄ laser system. This system not only serves as an effective method for

Figure 2. Schematic representation showing the principle of operation of a defect spot within a laser cavity to generate vortex laser emission. In a typical laser resonator, the laser beam output has a Gaussian intensity profile. By introducing an intra-cavity defect, we can break the symmetry of the resonator and force the oscillation of LG vortex modes.

generating vortex laser emission, but also as a laser which can be used to investigate non-linear wavelength conversion of vortex beams via the stimulated Raman scattering (SRS), and second harmonic (SHG) and sum-frequency generation (SFG) processes. There have been rather few studies of intra-cavity non-linear conversion of vortex laser beams, and the underlying dynamics of the transfer of topological charge under these processes have been little studied. The details of these non-linear conversion processes are discussed later in this chapter.

The end-pumped laser system investigated was comprised of an a-cut Nd:GdVO₄ crystal (0.3% Nd-doping), $4 \times 4 \times 20 \text{ mm}^3$ with a front surface coated with a high-reflecting coating ($R > 99.999\%$ at 1064 nm/ $R < 0.5\%$ at 880 nm) and an end-mirror with reflectivity $R = 99.91\%$ at 1064 nm and radius of curvature, 250 mm. Key here is that the end-mirror had an array of circular damage spots (3×3 array of spots with increasing diameter from 40–160 μm with a spacing of 400 μm) laser micro-machined (to the level of the glass substrate) onto its surface which acted as defect spots [21]. The laser cavity was end-pumped with a fibre coupled (100 μm diameter, 0.22 NA) laser diode producing up to 30 W at 879 nm. The experimental layout of the system is shown schematically in **Figure 3**, along with a schematic of the array of damage spots on the resonator mirror.

Alignment and optimisation of the laser system required accurate positioning of the end-mirror to ensure that the laser cavity mode was aligned with each defect spot. The operation of the system aligned on each defect spot was analysed, and it was found that complete suppression of the TEM₀₀ mode, and oscillation of the LG₀₁ mode could only be achieved with the 40 μm diameter defect spot. Given that the cavity mode had a diameter of $\sim 260 \mu\text{m}$, this corresponded to a cavity to defect ratio of 0.15, which was consistent with reports in the literature.

Figure 3. (a) Experimental layout of the diode-end pumped vortex laser utilising defect spots laser micro-machined onto the end-mirror [21]; and (b) the layout of the array of defect spots on mirror M2.

The threshold for oscillation of the LG_{01} mode from this system was achieved for an incident pump power of 0.1 W; maximum output power at 1064 nm was 0.4 W. The spatial profile of the laser emission was annular with a central dark spot, consistent with an LG_{01} mode. This spatial profile was retained across the pumping range. Beam quality factor (M^2) measurements of the output were made for incident pump powers of 0.1 W (just above laser threshold) and 2 W, and values of 2.2 and 3.1 were determined, respectively. An M^2 value of 2 is consistent with a vortex laser beam with topological charge of $\ell = 1$ [19]. The higher M^2 value at higher incident pump power is indicative of the onset of higher-order modes oscillating within the laser cavity, which is a consequence of the high laser gain and high cavity Q-factor.

The wavefront properties of the laser output were examined using an interferometric technique [4]. This technique yields a set of straight fringes, within which the position and order of the vortex beam phase-singularity can be determined through break-down in fringe structure- manifesting in a fork-like pattern. Both linear and spiral fringe patterns may be produced; in the case of a spiral fringe pattern, a spherical wavefront is formed using a short focal length lens [4]. Determination of the sign of the topological charge of the vortex beam using these methods relies on knowledge of the projection of the wavefront in the reference arm relative to the direction of propagation (in the case of straight fringe patterns); or knowledge of the radius of curvature of the reference beam in the case of spiral fringe patterns.

The spatial profile, along with the fork and spiral interferometer patterns for the laser output is shown in **Figure 4(a)–(c)**, respectively.

From **Figure 4(a)** it can be seen that an annular spatial profile is produced from this laser resonator, and from the interference patterns shown in **Figure 4(b)** and **(c)**, it can be seen that a single fork/spiral is observed, which is indicative of a phase singularity with a topological charge of 1. As the laser output was power scaled, the order of the topological charge and direction of rotation did not change. It was possible to alter the direction of the wavefront by altering the laser alignment through slight adjustment of the output coupler angle.

This result demonstrates the relative ease with which a vortex laser beam with a well-defined topological charge may be directly generated from an end-pumped solid-state laser system.

Figure 4. (a) Spatial profile of the 1063 nm emission from the laser resonator; and interferometer patterns showing (b) linear; and (c) spiral fringes [21].

This is significant as it precludes the requirement for additional vortex beam-shaping/forming components. As will be discussed further in this chapter, this simple resonator design facilitates investigation of intra-cavity non-linear wavelength conversion of vortex laser beams with relatively high efficiency.

2.2.2. Side pumped solid-state vortex laser

In a side-pumped laser resonator, the gain medium is pumped through a side facet as opposed to an end-face as described in the previous section. This has the effect of increasing the overall pump power that can be utilised in the system by distributing the thermal load throughout a larger area of the crystal [26]. In the side-pumped laser discussed in this section, the oscillating laser mode also undergoes reflection (total internal reflection) at the surface through which the pump radiation is delivered [24, 25]. In this configuration, oscillation of the vortex mode (LG_{01}) over the TEM_{00} (Gaussian mode) is achieved due to the thermal lens induced within the crystal. As the laser power is increased, the induced thermal lens affects the Gaussian mode faster (due to mode overlap considerations) and it is forced to be unstable (effectively being suppressed), while the LG_{01} vortex mode is still supported within the resonator. Therefore, this system operates with vortex output for a window of input pump powers, that is, enough pump power to induce a thermal lens sufficient to drive the system unstable for the Gaussian mode, while ensuring oscillation of the vortex mode. This allows for significantly high power vortex modes to be generated from this system.

The laser system comprises a laser diode side-pumped a-cut $Nd:GdVO_4$ slab (1% Nd-doping) with dimensions $2 \times 5 \times 20 \text{ mm}^3$, around which a bounce resonator is formed. The y - z faces of the slab were wedged and anti-reflection coated for 1063 nm to prevent self-lasing [26]. The laser mode oscillates between two flat mirrors, denoted E.M (high reflecting at 1063 nm) and O.C. (coated $R = 40\%$ at 1063 nm). To maintain good spatial overlap between the resonator mode and the pumped volume, two cylindrical lenses were used intra-cavity, denoted $CL_{1,2}$ both with focal length $f = 50 \text{ mm}$. The slab is side pumped by a laser diode bar array producing 55 W into an elliptical pump spot generated with a cylindrical lens ($f = 12.7 \text{ mm}$). The system layout is shown in **Figure 5**.

Figure 5. Layout of the side-pumped solid-state laser system [26].

Figure 6. Power transfer curve for the side-pumped bounce laser. Vortex beam emission is observed for incident pump powers in the range 43–55W [26].

The threshold for lasing action was ~ 10 W incident pump power. Above this threshold, the laser oscillates with a Gaussian-like output profile. As the incident pump power is increased, oscillation of an LG_{01} mode dominates and the TEM_{00} mode is suppressed. The power transfer curve for this system is shown in **Figure 6**. Spatial profiles of the output laser mode are shown in **Figure 7(a)–(c)** for a range of incident pump powers.

From this system, a vortex beam with 17.8 W of power at 1064 nm is generated for 55 W incident diode pump power. **Figure 7(d)** shows the spiral interference pattern of the vortex beam generated at an incident pump power of 55 W.

Figure 7. Spatial profile of the emission from the laser system operating at powers of (a) less than 43 W; (b) 43 W; (c) 55 W. The spiral interference pattern of the output generated at a pump power of 55 W is shown in (d) [26].

It was observed that the topological charge of the vortex beam did not change as the system was power scaled, maintaining a value of $l = 1$.

This work demonstrates that very high-power vortex laser beams can be generated directly from a laser system without the need for additional beam shaping components. By exploiting thermal lensing (a property often considered a problem in laser systems), vortex beams with very high output power can be generated directly from a solid-state laser.

3. Non-linear wavelength conversion of vortex laser beams

Non-linear wavelength conversion of laser beams is an efficient and effective method of altering the wavelength of laser beams, and these methods have been employed extensively in the case of Gaussian beams [27, 28]. Non-linear wavelength conversion within a non-linear crystal can generally be achieved through reaching a threshold power/intensity, in the case of $\chi^{(3)}$ processes; or phase matching in the case of $\chi^{(2)}$ processes. As will be discussed in this section, in these non-linear conversion processes, the conservation of energy and momentum offers a unique, interesting and powerful method of controlling not only the wavelength of the vortex beam, but also to control/manipulate the topological charge and resultant spatial profile of the beam.

In this section, we cover non-linear wavelength conversion of vortex laser beams under the processes of stimulated Raman scattering (SRS); sum-frequency and second-harmonic generation (SFG, SHG); optical-parametric oscillation (OPO), and difference-frequency generation (DFG). Through this analysis, we investigate the dynamics of vortex laser beams and how each of these non-linear optical processes, in conjunction with cavity design, allows us to achieve selective control of the topological charge of the vortex beam.

3.1. Stimulated Raman scattering (SRS)

SRS is a third-order non-linear process $\chi^{(3)}$ which can be used to convert the wavelength of a laser beam through the Raman scattering process [29–31]. It can be performed both for intra-cavity and extra-cavity configurations, and is most commonly achieved with the use of crystalline Raman-active media. In the SRS process, a fundamental laser field excites a Raman-active resonant mode (phonon) of the crystalline material. This leads to a coupling of energy and as a result, a photon with energy difference between the incident photon and the resonant mode is scattered; this is referred to as the Stokes field. In the case of stimulated Raman scattering, a resonant cavity is used to oscillate the Stokes field. In this process, the scattered Stokes photons stimulate the generation of more Stokes photons at the same wavelength/frequency, akin to stimulated gain within a laser medium [26]. This leads to rapid build-up of the Stokes photons from noise, with a well-defined intensity threshold for the process. The wavelength-shifts which can be achieved are therefore dependant on the Raman-active modes which exist within a material. There are a plethora of Raman-active crystalline materials which have been demonstrated effective at wavelength shifting via SRS; some of most commonly applied crystals (and their primary Raman shift) include: GdVO_4 (885 cm^{-1}), YVO_4 (892 cm^{-1}), KGdWO_4 (768 and 901 cm^{-1}) and BaWO_4 (926 cm^{-1}) [31].

In the case of conventional laser fields with Gaussian spatial distributions, SRS is a well-studied process both for intra-cavity and extra-cavity configurations. However, relatively few investigations of SRS conversion of vortex laser beams have been undertaken. In this section, we investigate the generation of vortex laser beams within the context of end-pumped solid-state lasers, as a continuation of work presented in the previous section on direct-generation of vortex laser beams using defect spots on end-mirrors.

The SRS process requires the generation of very intense fields to achieve threshold [31]. Therefore, in the case of Gaussian fields, the SRS process is often restricted to the very centre of the oscillating Gaussian mode, where the intensity of the beam is highest. This generally leads to the generation of Stokes beams which are of higher beam quality, owing to the Raman-beam-clean-up effect [32]. This is an important consideration as the spatial profile of the ‘fundamental’ beam has a direct impact on the resultant Stokes beam. In the case where a non-Gaussian field, for example, a vortex field, oscillates within the laser cavity, achieving SRS threshold is more difficult due to the annular spatial profile of the oscillating mode.

We examine here, wavelength-shifting of a fundamental field at $1063\text{--}1173\text{ nm}$ via intra-cavity SRS within a self-Raman $\text{Nd}:\text{GdVO}_4$ laser crystal. The experimental setup is identical to that shown in **Figure 3**; however, key here is that the mirrors M1 and M2 are coated to oscillate both the fundamental and Stokes fields, M1 with $R > 99.999\%$ at 1064 and 1173 nm , and M2

with $R = 99.91\%$ at 1064 nm and $R = 99.4\%$ at 1173 nm. The same array of damage spots were used as detailed previously [21].

The threshold for oscillation of the vortex Stokes field at 1176 nm was ~ 2 W absorbed diode pump power, and a maximum Stokes power of 380 mW was generated from this laser for an absorbed pump power of 6.8 W. It was found that the Stokes beam retained an annular profile throughout the input pump power range. The spatial profile of the Stokes beam along with both linear and spiral interference patterns are shown in **Figure 8(a)–(c)**, respectively.

The annular spatial profile of the Stokes beam is a result of the SRS gain profile being dictated by that of the fundamental field mode. As the fundamental field is an LG_{01} mode, the SRS gain profile will also be annular in profile. Furthermore, the Stokes field is also affected by the damage spot laser machined onto the output mirror (M2). These two factors ensure that the Stokes field will retain an annular profile similar to that of the fundamental field.

The magnitude and sign of topological charge was always found to be the same for both the Stokes and fundamental fields. Here, it is interesting to consider what also happens to the phonon field which is also excited in the SRS process. Due to conservation of momentum rules, it is also possible for this field to receive topological charge from the fundamental field. To investigate this, we can consider conservation of orbital angular momentum under the process of SRS.

Let us denote the topological charge of the exciting fields as l_F , l_S and l_R for the fundamental, Stokes and material, respectively; and l'_F , l'_S , l'_R as the stimulated fields for the fundamental, Stokes and material, respectively. In a stimulated scattering event, the stimulated field must take on the same energy state as the stimulating field, so in this case, $l_S = l'_S$. Then in the stimulated scattering process, $l_F + l_S + l_R = 2l'_S + l'_R$.

We have observed that $l_F = l_S$ and so, there is no change in the topological charge state of the medium, that is, $l_R = l'_R$, that is, the material phonon does not receive orbital angular

Figure 8. Images of (a) the spatial profile of the output Stokes beam; (b) linear interference pattern; and (c) spiral interference pattern [21].

momentum in the SRS process. It is interesting to note that while transfer of topological charge from photons to phonons was not observed in this work under the process of SRS, it has been reported in the literature through non-linear Brillouin interactions [33].

This work demonstrates how readily the wavelength of a vortex laser beam can be converted using the SRS process; in this case, a fundamental wavelength at 1063 nm is converted to 1173 nm in GdVO_4 . Given the broad range of Raman-active crystalline materials that are available, the ability to generate new vortex laser wavelengths through the SRS process with the retention of topological charge state is very powerful.

3.2. Sum-frequency generation and second harmonic generation

Sum-frequency generation and second harmonic generation (an example of sum-frequency generation where two photons of the same frequency are combined) are effective non-linear methods of decreasing the wavelength of laser fields [28]. It is most commonly used to convert wavelengths in the near-infrared to the visible region. For example, SHG of 1064 nm (Nd:YAG emission) is commonly used to generate green emission (532 nm). This is the non-linear process employed in the now ubiquitous green laser pointer.

SFG is a phase-matched, non-linear process and requires the conservation of both momentum and energy. As already discussed in the context of SRS, it is this conservation of momentum which imparts special relevance to vortex laser fields in that orbital angular momentum (OAM) must be conserved. In the case where two vortex beams with topological charge l_{ω_1} and l_{ω_2} are transformed under SFG, the resultant field must have topological charge which is the sum of the two initial fields, i.e. $l_{\omega_1+\omega_2} = l_{\omega_1} + l_{\omega_2}$.

Through this process, we have a powerful means of manipulating both the wavelength and the topological charge of the vortex laser beam. The process of second harmonic generation of vortex beams has been studied comprehensively in the context of extra-cavity conversion [34, 35], however, this process in an intra-cavity configuration is less well understood. In this work, we investigate this process in the context of an intra-cavity, end-pumped Raman laser, as detailed in the above section for SRS. In this case, the SFG/SHG crystal is incorporated into the laser cavity, and the process of SFG or SHG is achieved through appropriate phase matching. The resonator layout is shown in **Figure 9**.

Figure 9. Layout of the intra-cavity vortex laser incorporating both SRS and SFG/SHG non-linear processes [22, 23].

The non-linear crystal used for SFG/SHG is a lithium triborate (LBO) crystal, cut for non-critical phase matching (NCPM), with dimensions, $4 \times 4 \times 10 \text{ mm}^3$. The LBO crystal was placed in a copper mount which could be temperature-controlled, to enable selective phase matching via temperature tuning. With the laser generating a fundamental wavelength of 1063 nm and a Stokes wavelength of 1173 nm, phase matching for SHG of the 1173 nm wavelength to 586 nm is achieved with the LBO crystal set to a temperature of 43.5°C , and SFG of the 1063 nm and 1173 nm fields is achieved at 93°C to generate output at 559 nm.

With this system, a maximum power of 727 mW at 586 nm, and 850 mW at 559 nm was achieved [22, 23]. The spatial profile of the SHG and SFG fields were similar to the annular profiles that were produced in the near-field, however these evolved to a spot with a central bright core in the far-field. The spatial profiles of the 586 nm and 559 nm fields in the near- and far-field are shown in **Figure 10**.

The beam quality-factor of the resultant beams were also measured. In the case of the near-field profiles, beam quality factors of $M^2 \sim 3\text{--}3.6$ were determined in both cases. A beam quality factor of $M^2 = 3$ is consistent with a vortex beam having topological charge $l = 2$.

The wavefront properties of the SFG and SHG beams were also analysed using the interferometric setup already described. Observations of the topological charge of the resultant SFG and SHG fields were consistent with conservation of orbital angular momentum, where the topological charge of the resultant field was always the sum of the topological charge of the initial fields, that is, $l_{\text{SFG}} = l_{\omega 1} + l_{\omega 2}$, wherein the degenerate case of SHG, $l_{2\omega} = 2l_{\omega}$. A comparison of the linear and spiral interference patterns of the Stokes (1173 nm) field and its SHG (586 nm) are shown in **Figure 11**.

The interference patterns clearly show that the Stokes field has a topological charge of $l = 1$, and the SHG field has a topological charge of $l = 2$ (three-pronged fork and two spirals). This shows that doubling of the topological charge takes place under SHG, further supporting the near-field spatial profile with a large central dark region as shown in **Figure 10(a)**.

In the case of SFG of the 1063 nm and 1173 nm fields, two different interference patterns were observed, one consistent with the generation of an SFG vortex field with topological charge

Figure 10. Spatial profiles of the 586 nm (SHG) field in the (a) near-field; and (b) far-field. Spatial profile of the 559 nm (SFG) field in the (c) near-field; and (d) far-field [22, 23].

Figure 11. Comparison of the interference patterns of the Stokes (1173 nm) field (a) linear; and (b) spiral interference patterns, with that of the SHG field (586 nm) (c) linear; and (d) spiral interference patterns [22].

$l = 2$, and the other consistent with an SFG field with topological charge of $l = 0$. It should be noted that in both cases, the same annular, near-field profile is generated. The linear interference patterns showing these two conditions are shown in **Figure 12**.

The two different topological charge states of the SFG field results from two different topological charge states of the 1063 nm and 1173 nm fields. In the case where the SFG field has a topological charge $l = 2$, both the 1063 nm and 1173 nm fields have topological charge $l = 1$. In the case where the SFG field has a topological charge of $l = 0$, the 1063 nm and 1173 nm fields have opposite sign of topological charge, that is, $l = +1$ and $l = -1$ or vice-versa. It is interesting to note that in this resonator configuration, where an LBO crystal is incorporated cf. that where only SRS takes place, it is possible to achieve a situation in which the 1063 nm and 1173 nm fields have opposite topological charge. The ability of the Stokes field to take on opposite topological charge to that of the fundamental field is likely due to the additional non-linear process of SFG competing with the SRS process in depleting the fundamental field.

Figure 12. Interference patterns generated from the SFG of 1163 nm and 1173 nm fields, showing (a) topological charge of 2; and (b) topological charge of 0 [23].

This generates instability within the system which affects mode symmetry, and enables oscillation of opposite topological charge.

What is significant in these results is that an SFG field can be generated with topological charge 0 and yet retain an annular spatial profile in the near-field. By definition, a vortex beam with zero topological charge should have a non-zero central core. Also of significance is that the spatial profile of the SHG and SFG fields is annular in the near-field, and develop a central bright spot in the far-field. This very interesting beam dynamics can be understood through a decomposition of the oscillating LG₀₁ modes at 1063 nm and 1173 nm into their constituent Hermite-Gaussian (HG) modes, and examining how these modes evolve under the process of SFG/SHG and under free-space propagation. This process has been computationally modelled for comparison with experimental results.

In this model we first assume that the LG resonator modes for the fundamental and Stokes fields comprise Hermite-Gaussian (HG) modes, HG_{1,0} and HG_{0,1} where,

$$HG_{1,0} = x e^{-(x^2+y^2)} \quad (1)$$

$$HG_{0,1} = y e^{-(x^2+y^2)} \quad (2)$$

The expressions for the Laguerre-Gaussian vortex beams of the fundamental field with frequency (ω_1) and the Stokes field with frequency (ω_2) are given by:

$$LG_1^{\omega_1} = x e^{-(x^2+y^2)} + iy e^{-(x^2+y^2)} e^{i\Delta_1} \quad (3)$$

$$LG_{\pm 1}^{\omega_2} = x e^{-(x^2+y^2)} \pm iy e^{-(x^2+y^2)} e^{i\Delta_2} \quad (4)$$

where Δ_1 and Δ_2 are small phase mismatch terms which can manifest within the resonator [21]. In the expression for the Stokes field, ± 1 denotes that the Stokes field can take a topological charge value of +1 or -1.

Now in the case of second harmonic generation, let us simply consider the case of SHG of ω_1 . Here, the field strength of the SHG field is given by:

$$\begin{aligned} E_{SHG} &= LG_1^{\omega_1} \cdot LG_1^{\omega_1} = (x^2 - y^2 e^{2i\Delta_1}) e^{-2(x^2+y^2)} + 2ixy e^{i\Delta_1} e^{-2(x^2+y^2)} \\ &= HG_{2,0} - HG_{0,2} e^{2i\Delta_1} + \frac{1}{2}(1 - e^{2i\Delta_1})HG_{0,0} + 2i e^{i\Delta_1} HG_{1,1} \end{aligned} \quad (5)$$

where $HG_{2,0} = (x^2 - \frac{1}{2})e^{-2(x^2+y^2)}$, $HG_{0,2} = (y^2 - \frac{1}{2})e^{-2(x^2+y^2)}$, $HG_{1,1} = x \cdot y \cdot e^{-2(x^2+y^2)}$

Note that in the above equation, the Gaussian term, in the form HG_{0,0} manifests. This term does not have a central dark core in its spatial profile. Under propagation along an axis z , the contribution of Gouy phase on the modes as they propagate must also be considered. For HG modes, the Gouy phase term is represented as:

$$\delta_{m,n} = (m + n + 1) \tan^{-1} \frac{z}{z_R} \quad (6)$$

where m and n are the indices of the HG mode with the form HG _{m,n} , z is the propagation distance, and z_R is the Rayleigh length. For the HG modes under consideration here,

$$\delta(z) = \delta_{2,0} - \delta_{0,0} = \delta_{0,2} - \delta_{0,0} = \delta_{1,1} - \delta_{0,0} = 2 \tan^{-1} \frac{z}{z_R}$$

The intensity of the SHG field is given by,

$$I_{SHG}(z) = |E_{SHG}|^2$$

$$I_{SHG}(z) = |HG_{2,0}|^2 + |HG_{0,2}|^2 + 4|HG_{1,1}|^2 + \Delta_1^2 |HG_{0,0}|^2 - 2(1 - 2\Delta_1^2)HG_{2,0}HG_{0,2} + 2\Delta_1^2 \cos(\Delta\delta) (HG_{2,0} + HG_{0,2})HG_{0,0} \quad (7)$$

where for small Δ_1 $\langle \sin \Delta_1 \rangle \simeq 0$, $\langle \cos \Delta_1 \rangle \simeq 1$, $\langle \sin^2 \Delta_1 \rangle \simeq \Delta_1^2$

$$\delta(z) = \delta_{2,0} - \delta_{0,0} = \delta_{0,2} - \delta_{0,0} = \delta_{1,1} - \delta_{0,0} = 2 \tan^{-1} \frac{z}{z_R}$$

From Eq. (7) we can see that Gouy phase impacts the spatial intensity profile of the SFG field in the far-field as its finite value (as Z increases) means that there is contribution to the overall spatial profile by the $HG_{0,0}$ mode. Following this derivation, we can also form expressions for the SFG field under conditions where the fundamental ω_1 and Stokes ω_2 fields have the same and opposite topological charge. The expressions for the intensity of the SFG fields in each case are given by:

$$I_{SFG}^{l=2}(z) = |HG_{2,0}|^2 + |HG_{0,2}|^2 + (\Delta_1 + \Delta_2)^2 |HG_{0,0}|^2 + 4|HG_{1,1}|^2 - 2HG_{2,0}HG_{0,2} + \frac{(\Delta_1 + \Delta_2)^2}{2} \cos\delta (HG_{2,0} + HG_{0,2})HG_{0,0} \quad (8)$$

$$I_{SFG}^{l=0}(z) = |HG_{2,0}|^2 + |HG_{0,2}|^2 + |HG_{0,0}|^2 - (\Delta_1 + \Delta_2)^2 |HG_{1,1}|^2 + 2HG_{2,0}HG_{0,2} + 2\cos\delta (HG_{2,0} + HG_{0,2})HG_{0,0} \quad (9)$$

The spatial profile for the SHG field and the SFG field under the two different conditions where the fundamental and Stokes fields have the same, and opposite topological charge have been simulated, and are shown in **Figure 13**.

The simulated spatial intensity profiles of the SHG and SFG beams in the near- and far-fields very well replicate the spatial profiles observed experimentally in **Figure 10**. The interference patterns of the SFG fields under the conditions where the fundamental and Stokes have the same topological charge, and the opposite topological charge were simulated, and these are shown in **Figure 14(a)** and **(b)**, respectively.

The simulated interference patterns clearly replicate the experimental results and show that while the SFG field can have an annular spatial intensity profile in the near-field, it is also possible that the beam does not have a finite topological charge, and exhibits a topological charge $l=0$ (in this case, occurring where fundamental and Stokes beams have opposite topological charge).

Figure 13. Simulated spatial intensity profiles of the SHG beam in (a) near-field; and (b) far-field. Simulated spatial intensity profile of the SFG field in (c) near-field; and (d) far-field, where the fundamental and Stokes fields have the same topological charge. Simulated spatial intensity profile of the SFG field in (e) near-field; and (f) far-field, where the fundamental and Stokes fields have opposite topological charge [22, 23].

This study of vortex beam dynamics under SHG and SFG within a laser cavity has yielded interesting results. Under the process of SHG or SFG, the wavelength of the vortex beam is converted as expected. The spatial intensity profile in the near-field is that of a vortex beam with a central dark spot with a size consistent with a vortex beam with the magnitude of topological charge equal to the sum of the magnitude of topological charge of the initial fields. What was unexpected was the evolution of the spatial intensity profile of the SHG/SFG vortex field as it propagates out of the laser cavity. While in the near-field, a perfect annular spatial

Figure 14. Simulated interference patterns for the SFG field where (a) both fundamental and Stokes fields have the same topological charge; and (b) fundamental and Stokes fields have opposite topological charge [23].

intensity profile is produced, as it propagates into the far-field, the profile evolves to resemble that of an annular beam with a central bright spot. Through decomposition of the vortex modes constituting the fundamental and Stokes modes, into HG modes, it is revealed that upon propagation out of the laser resonator, the contribution of Gouy phase shift must also be considered. This Gouy phase directly impacts the spatial profile of the vortex beam in the far field, resulting in the presence of a non-zero central region.

The variation in the spatial profile of the vortex beam in the near- and far-fields is interesting as this adds to the flexibility and range of applications for this type of laser beam. In one instance, the annular profile in the near-field may be re-imaged for applications where a central null is required, however in other applications, the far-field profile can be utilised where a Gaussian-like beam is required. Furthermore, it has been demonstrated that under SFG, it is possible to produce an annular beam without a net topological charge. This may also be of use in certain applications where an annular spatial profile is desired, but orbital angular momentum is not.

3.3. Optical parametric oscillation

OPO is an effective means of significantly increasing the wavelength of a laser beam. Using OPOs, laser beam wavelengths can be extended from the visible out to the infra-red [28, 36, 37]. The efficiency with which this can take place is dependent on the effective non-linearity of the non-linear crystal being used for conversion, and its transparency in the wavelength range being generated. In this non-linear process, an initial laser field, designated the ‘pump’ is split into two laser fields of lower energy (longer wavelength). The field with the longer wavelength is designated the ‘idler’, while that of the shorter wavelength, the ‘signal’. The sum of the frequency of the signal and idler equal that of the pump, that is, $\omega_i + \omega_s = \omega_p$.

The dynamics of vortex laser beam transformation under optical parametric oscillation has been studied in the context of continuous-wave OPOs [38, 39]; however in these studies, the wavelength tuning diversity and power scaling properties of vortex beams produced in this non-linear process has not been examined. In this section, we examine the process of topological charge transfer and wavelength extension of vortex laser beams via the OPO process, with pump beams initially at wavelengths of 1064 and 532 nm. Conventional solid-state lasers producing nanosecond pulsed radiation at these wavelengths are used, along with spiral phase plates (see Section 2.1) to transform these Gaussian laser beams into vortex laser beams with topological charge $l = 1$. This vortex beam (pump field) is then injected into an optical parametric oscillator and converted to the signal and idler fields. By altering the resonator geometry and the crystal used for the OPO process, the dynamics by which the pump vortex beam is transformed, to generate signal and idler fields with different topological charge can be controlled. By tuning the phase matching conditions within the OPO, significant wavelength tuning of the generated signal and idler fields can be achieved.

Four different OPO setups are discussed here. The general OPO system layout is depicted in **Figure 15**, and the properties of each element used in the four configurations are summarised in **Table 1**.

Figure 15. General vortex OPO system layout.

For each of the resonator configurations listed in Table 1, the topological charge transfer process from the pump to the signal and idler fields is different, and this is detailed in the following sections.

3.3.1. Plane-plane OPO resonator using KTP (Setup A)

In this system, a KTP crystal is used to phase match for degenerate OPO operation, in which the wavelength of the signal and idler fields are the same. The incident pump field is a vortex laser beam with topological charge $l = 1$ at a wavelength of 1064 nm, and the resultant signal

Setup	A (plane-plane OPO using KTP)	B (concave-concave OPO using KTP)	C (concave-plane OPO using cascaded KTP)	D (plane-concave OPO using cascaded LBO)
Pump laser	1064 nm, 45 ns, 50 Hz	1064 nm, 25 ns, 50 Hz	1064 nm, 25 ns, 50 Hz	532 nm, 25 ns, 50 Hz
Spiral phase plate	Designed to produce $\ell = 1$ vortex at 1064 nm	Designed to produce $\ell = 1$ vortex at 1064 nm	Designed to produce $\ell = 1$ vortex at 1064 nm	Designed to produce $\ell = 1$ vortex at 532 nm
Focusing lens/spot diameter on OPO crystal	$f = 700$ mm lens	Focal spot = 520 μm diameter	Focal spot = 450 μm diameter	Focal spot = 750 μm diameter
Input mirror	Flat, $R = 98\%$ at 2 μm ; $T = 90\%$ at 1 μm	Radius of curvature (ROC) = 2000 mm, $R = 98\%$ at 2 μm ; $T = 90\%$ at 1 μm	Radius of curvature (ROC) = 2000 mm, $R = 98\%$ at 2 μm ; HT at 1 μm	Flat, HR at 980 nm, HT at 532 nm
OPO crystal	KTP, $5 \times 5 \times 30$ mm ³ , $\theta = 51.4^\circ$	KTP, $5 \times 5 \times 30$ mm ³ , $\theta = 51.4^\circ$	Two cascaded KTP crystals, $12 \times 9 \times 27$ mm ³ , $\theta = 53^\circ$	Two cascaded LBO crystals, 30×33 mm ³ , $\theta = 90^\circ$, $\phi = 0$
Output mirror	Flat, $R = 80\%$ at 2 μm ; HT at 1 μm	ROC = 100 mm, $R = 80\%$ at 2 μm ; $T = 80\%$ at 1 μm	Flat, $R = 50\%$ at 2 μm ; HT at 1 μm	Flat folding mirror with HR at < 980 nm and HT at > 1180 nm; combined with either: concave mirror, $R = 80\%$ at 980 nm (resonate signal) OR concave mirror, $R = 60\%$ at 1180 nm (resonate idler)

Table 1. Details of each resonator element used in the different OPO configurations examined in Refs. [40–43].

and idler fields have the same wavelength of 2128 nm. In this system, the topological charge of the pump beam was observed to evenly split between the signal and idler fields, in this case, they both receive a topological charge of $l = \frac{1}{2}$. Spatial profiles of the pump, signal and idler fields are shown in **Figure 16**.

It can be clearly seen that the spatial profile of the signal beam (**Figure 16(b)**) resembles a half-crescent shape, while that of the idler (**Figure 16(c)**) appears somewhat Gaussian. The topological charge state of the signal and idler fields is determined through frequency doubling of the fields. Here, the spatial profiles of the frequency-doubled signal and idler are shown in **Figure 17(a)** and **(b)**, respectively; and their corresponding interference patterns in **Figure 17(c)** and **(d)**, respectively.

The spatial profile and interference pattern (**Figure 17(a)** and **(c)**) of the frequency-doubled signal beam resembles that of a vortex beam with a topological charge of 1, confirming that the signal field has a topological charge of $\frac{1}{2}$, knowing that the topological charge of the frequency-doubled field is double that of the original field (see Section 3.2). The spatial profile of the frequency-doubled idler shows an offset null region (**Figure 17(b)**); this offset is due to the effect of beam-walk-off for the polarisation of the idler beam (o-wave) in this OPO phase matching geometry. From the interference pattern (**Figure 17(d)**), it is clear that this beam also shows a topological charge of 1, also showing that the idler field also has a topological charge of $\frac{1}{2}$. In this system, isotropic sharing of the topological charge of the incident pump field between the signal and idler takes place.

3.3.2. Concave-concave OPO resonator using KTP (Setup B)

In this system, in contrast to the plane-plane resonator, the resonator mirrors are both concave. The same KTP crystal was used to phase match the OPO process. In this system, wavelength tuning of the signal and idler fields was also investigated by changing the phase matching conditions.

It was found that the signal field could be tuned through a wavelength range of 1953–2158 nm [41]. Interestingly, in this system, isotropic topological charge sharing from the pump to signal and idler fields was not observed. Instead, anisotropic charge transfer occurred

Figure 16. Spatial intensity profiles of (a) pump; (b) signal; (c) idler fields generated from the plane-plane OPO resonator using KTP in a degenerate phase matching configuration [40].

Figure 17. Spatial intensity profiles of (a) frequency-doubled signal, (b) frequency-doubled idler field, and interference patterns of (c) the frequency-doubled signal field, and (d) the frequency-doubled idler field [40].

between the pump and signal fields. In this case, the signal field always received the topological charge of the pump field, whilst the idler field remained Gaussian with topological charge $\ell = 0$. Images of the spatial profile of the pump, signal and idler fields are shown in **Figure 18(a)–(c)**, respectively. The topological charge state of the signal and idler beams was directly determined (not frequency-doubled as detailed in Section 3.3.1) through the

Figure 18. Spatial intensity patterns for (a) pump, (b) signal and (c) idler fields. The interference pattern generated through a self-referencing technique is shown for (d) the signal and (e) the idler fields [41].

use of a self-referencing interference technique [41]. In this technique, a set of two fork-like patterns are generated instead of one as generated through the Mach-Zehnder interferometer detailed previously. The interference patterns for the signal and idler fields are shown in **Figure 18(d)** and **(e)**, respectively. The fork pattern seen in **Figure 18(d)** shows that the signal field has a topological charge $\ell = 1$, the same as the input pump beam, and the absence of any fork pattern in **Figure 18(e)** shows that the idler field has a topological charge $\ell = 0$.

The anisotropic topological charge transfer which occurs in this system can be understood through examination of how the signal and idler beams are produced within the OPO resonator. In contrast to the plane-plane resonator, the plane-concave resonator has a finite Rayleigh range of $Z_R = 50\text{mm}$. Due to this property, the effect of Gouy phase on the signal and idler fields must be considered, similar to that already discussed in the context of intracavity SFG/SHG. It is also important to realise that the pump and signal fields have the same polarisation (both e-waves), while the idler has an orthogonal polarisation (o-wave) due to the OPO phase matching condition used in the KTP crystal (type-II phase matching). Due to the finite Rayleigh length, there exists a phase shift between a Gaussian mode and the $\ell = 1$ vortex mode of ~ 1.2 rad. This effectively means that the overlap between a Gaussian mode and the vortex mode is reduced. Furthermore, the idler field also exhibits beam walk-off in the KTP crystal due to its polarisation state. The net effect of this walk-off is that the overlap between the idler mode and the vortex pump mode is impacted, significantly reducing the possibility of the idler mode taking on the profile of the vortex pump field. It is these two factors, Gouy phase shift and beam walk-off, within this OPO resonator configuration, which enforces topological charge transfer from the pump field to the signal field, while the idler field remains Gaussian.

With this concave-concave resonator design, the use of two KTP crystals simultaneously in a cascaded configuration was also investigated (**Setup C**) [42]. This enabled the wavelength tuning of both the signal and idler waves, and compensation of walk-off of the idler beam. Using this configuration, wavelength tuning of the signal beam across the range 1820 - 1954 nm and tuning of the idler beam across the range 2561–2335 nm could be achieved.

3.3.3. Plane-concave resonator using LBO (*Setup D*)

In this system a plano-concave OPO resonator was used, however, the KTP crystal was replaced for lithium niobate (LBO) cut for non-critical (type I) phase matching. In this phase matching scheme, an e-wave is converted into two o-waves $e \rightarrow o + o$. In this setup, two cascaded LBO crystals were used to enable wavelength tuning of both the signal and idler fields. Also, in this system, the pump field was generated at 532 nm instead of 1064 nm as used in the previous setups.

Initially, this system was setup to resonate both the signal and idler fields in a doubly resonant configuration. Due to a lack of walk-off of the signal or idler fields (due to the LBO crystal and phase matching geometry), no preferential topological charge transfer was observed from the pump to either of these fields. As a result, the signal and idler fields were both observed to exit the OPO resonator with mode structure which is an incoherent superposition of both

Gaussian and vortex mode profiles. To achieve preferential topological charge transfer from the pump field to either the signal or the idler field, it was necessary to resonate only one of these fields. In the case where only the signal field was oscillated, the topological charge of the pump was transferred to the signal, and similarly if only the idler field was resonated, the topological charge of the pump was transferred to the idler field. Hence, anisotropic topological charge transfer from the pump to the signal or idler fields could be selectively achieved. Wavelength tuning of the signal could be achieved across a range 850–990 nm, and that of the idler from 1130–1300 nm [43].

3.4. Difference-frequency generation of vortex beams

In addition to wavelength extension of vortex beams using an OPO, further wavelength extension of these vortex beams was explored by using DFG of the generated signal and idler fields. In this work, the signal and idler output from the plano-concave resonator incorporating cascaded KTP crystals (Setup C) was focussed into a zinc germanium phosphide (ZGP) crystal for DFG conversion under Type-I phase matching. It was found that through tuning of the signal and idler wavelengths, the resultant DFG field could be wavelength tuned through a range 6.3–12 μm , significantly extending the wavelength reach of vortex laser beams into the mid/far infra-red [42].

In this configuration, it was also found that conservation of topological charge was maintained in this process. In the DFG process, the conservation law of topological charge follows the relationship:

$$l_{\text{DFG}} = \frac{\omega_s - \omega_i}{|\omega_s - \omega_i|} (l_s - l_i) \quad (10)$$

where ω_s and ω_i are the frequencies of the signal and idler fields, respectively, and l_{DFG} , l_s and l_i are the topological charge of the DFG, signal and idler fields, respectively. From Eq. (10), it can be seen that the sign of the topological charge of the DFG field is dependent on the frequencies of the signal and idler fields. When the frequency of the signal is greater than that of the idler, a positive topological charge is produced, and when the frequency of the idler is greater than the signal, a negative topological charge is generated. Therefore, by swapping the frequency of the signal and idler fields, it is possible to swap the sign of the topological charge of the resultant DFG field, thereby imparting an additional method of controlling the topological charge state of the generated vortex field.

From these studies, it is clear that OPOs offer an effective method of significantly extending the wavelength diversity of vortex laser beams. Interestingly, the dynamics by which the beams are converted can be readily controlled through careful consideration of the OPO resonator design and the crystals which are used in the OPO process. Here, not only can control of the wavelength of the vortex beam be achieved, but also, the transfer of topological charge of the pump beam to the signal and idler beams can be manipulated. Significantly, the generation of a vortex laser beam with $l = \frac{1}{2}$ can be achieved through this non-linear conversion process. In contrast to conventional vortex laser beams with an annular spatial profile, this crescent spatial profile lends itself to numerous new applications including fabrication of novel devices such as meta-materials.

3. Summary

In this chapter, we have presented results detailing both the direct generation of vortex laser beams from solid-state laser systems, and their wavelength conversion via non-linear optical methods. The results show the great diversity of vortex outputs that can be generated, both in terms of spatial profile, topological charge and wavelength. It is through this continual examination of the dynamics of vortex laser beams, that the characteristics of these beams can be manipulated, and thus expanding the range of applications to which they can be applied.

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