Cascade Raman sideband generation and orbital angular momentum relations for paraxial beam modes

J. Strohaber,1,* J. Abul,2 M. Richardson,1 F. Zhu,2 A. A. Kolomenskii,2 and H. A. Schuessler2

1Department of Physics and Astronomy, Florida A&M University, Tallahassee, FL, 32307, USA
2Department of Physics and Astronomy, Texas A&M University, College Station, TX, 77843-4242, USA

*james.strohaber@famu.edu

Abstract: In this work, the nonlinear parametric interaction of optical radiation in various transverse modes in a Raman-active medium is investigated both experimentally and theoretically. Verification of the orbital angular momentum algebra (OAM-algebra) [Strohaber et al., Opt. Lett. 37, 3411 (2012)] was performed for high-order Laguerre Gaussian modes \( l > 1 \). It was found that this same algebra also describes the coherent transfer of OAM when Ince-Gaussian modes were used. New theoretical considerations extend the OAM-algebra to even and odd Laguerre Gaussian, and Hermite Gaussian beam modes through a change of basis. The results of this work provide details in the spatiotemporal synthesis of custom broadband pulses of radiation from Raman sideband generation.

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1. Introduction

Optical beam modes are transverse eigensolutions of the paraxial wave equation (PWE) of electrodynamics [1]. The modes hold for beams that are well collimated such that the curvature in the propagation direction can be neglected. Three families of solutions have been found by separation of variables and take on the geometry of their respective coordinate system such as Cartesian, cylindrical and elliptical polar coordinates [1, 2]. The fundamental mode in all three families is the commonly encountered Gaussian beam produced by many laser systems. Of these solutions, optical vortices have attracted considerable attention from the scientific community because they possess an external degree of freedom known as optical orbital angular momentum (OAM) in addition to the internal spin angular momentum [3]. Applications of optical modes include: optical manipulation of Bose Einstein (BECs) condensates, such as tight confinement using Hermite Gaussian (HG) modes [4]; transfer of angular momentum with Laguerre Gaussian (LG) modes [5, 6]; optical spanners [7] and pumps [8]; imaging of cytoskeletal filaments (stimulated emission depletion) [9] multiplexing and cryptography [10, 11] and the search for exoplanets (optical vortex coronagraph) [12]. The success of many high-field experiments is possible by advances in the generation and analysis of beam modes in femtosecond fields [2, 13, 14]. However, investigations of highly nonlinear interactions of beam modes with matter have remained largely unexplored.

Recently a number of investigations involving the nonlinear interaction of optical vortices with matter appeared in the literature [15—17]. In experiments presented in [15], the authors crossed time-delayed chirped pulses in a lead tungsten PbWO4 crystal and demonstrated for the first time the coherent transfer of optical angular momentum in the generation of cascaded Raman sidebands. In these experiments, topological charge measurements verified a predicted orbital angular momentum algebra (OAM-algebra). This OAM-algebra was later verified in experiments given in [16, 17]. In [16] the authors employed two optical parametric amplifiers to produce two spectrally separated beams that were crossed in a Raman active crystal, and in [17] the authors utilized a dichroic beam splitter and spectral filters. In each of these previous experiments, a spiral phase plate was used producing only the \( \ell = \pm 1 \) transverse mode. This therefore limited the scope of the previous experiments.

In the present work, we extend upon our earlier work [15] by investigating the production of Raman sidebands with higher-order helical beams such as the Laguerre Gaussian (LG) and Ince Gaussian (IG) modes; the non-helical even Laguerre Gaussian (LG\(^\pm\)) and odd Laguerre Gaussian (LG\(^\mp\)), and Hermite Gaussian modes (HG). From our experiments, we verify that the OAM-algebra holds for the high order helical-LG beams as well as for the helical-IG modes. In the case of the non-helical modes, such as the even and odd LG modes or the HG modes, the OAM-algebra derived in [15] cannot be used; therefore, a new OAM-algebra is developed that describes the quantum coherent transfer of phase and amplitude information into the sideband orders.
Throughout this work all optical beam modes were produced using computer generated holograms (CGH) displayed on a liquid crystal spatial light modulator (SLM). The CGHs were produced by interfering a plane wave with the paraxial solutions of the desired beam mode. These solutions, which correspond to different coordinate geometries are:

\[
HG_{m,n} = N_{mn} H_n \left( \frac{\sqrt{2} x}{w} \right) e^{-\frac{x^2}{2w^2}} H_m \left( \frac{\sqrt{2} y}{w} \right) e^{-\frac{y^2}{2w^2}} e^{i(kx+ly+\omega t+\varphi_j)}, \quad (1a)
\]

\[
LG_{\rho,\ell}^{e(o)} = N_{\rho}\rho \left( \frac{\sqrt{2}r}{w} \right) l_{\rho} \left( \frac{2r^2}{w^2} \right) \left( \cos(\ell \theta) \right) \left( \sin(\ell \theta) \right) e^{-\frac{r^2}{w^2}} e^{i(2\rho \ell+1\ell+\varphi_j)}, \quad (1b)
\]

\[
IG_{\rho,m}^{e(o)} = N_{mn} \left( \frac{C_{\rho}(i\xi,e)}{S_{\rho}(i\xi,e)} \right) e^{-\frac{r^2}{w^2}} e^{i(2\rho \ell+1\ell+\varphi_j)}, \quad (1c)
\]

Here \(HG\), \(LG\), and \(LG\)’ are as mentioned earlier, and \(LG\’\) (\(IG\’\)) are the even (odd) Ince Gaussian beams. The helical LG and IG beams are found by the complex superposition of the even and odd solutions having the same mode numbers [18], i.e., \(LG_{\rho,\ell} = LG_{\rho,\ell}^{e} + \text{sgn}(\ell)LG_{\rho,\ell}^{o}\) and \(IG_{\rho,m} = IG_{\rho,m}^{e} \pm IG_{\rho,m}^{o}\) [18]. The polynomials used are: the Hermite [14], Laguerre [14], as well as the even and odd Ince [18] respectively. Reference [18] contains the full derivation of the IG modes.

2. Experimental setup

In our experiments, amplified radiation from a Ti:sapphire laser system was produced by chirped-pulse amplification (Coherent Legend) producing ~50 fs laser pulses at a repetition rate of 1 kHz. The central wavelength was 800 nm, and the energy per pulse was ~1 mJ. Output radiation from the laser was sent through a variable iris to control both mode quality [16] and beam power in the setup. Following the iris, the radiation was sent through a four-port Michelson interferometer Fig. 1(a). The purpose of the four-port interferometer is to produce a beam encoded with the phase information of the desired optical mode and to produce a reference beam needed to investigate the phase content of the generated sidebands. In contrast to our previous setup [15], one of the mirrors in the stationary arm of the four-port was replaced by a parallel aligned liquid crystal on silicon spatial light modulator (Hamamatsu LCOS-SLM 104683). This device was addressed by a control unit and computer to display the CGHs. The inset in Fig. 1(a) shows two examples of such holograms. A routing mirror was used to steer radiation from the output of the four-port Michelson into the beam crossing setup shown Fig. 1(b). A lens (L) having a nominal focal length of 40 cm was used to focus the pump and Stokes beams into the Raman-active crystal. The separation distance between the beams at the position of the lens was ~2.25 cm and resulted in a full-angle of 3.2 degrees between the two beams. The nonlinear crystal was a 0.5 mm thick lead tungsten crystal (MTI Corporation). Images of the pump and Stokes beams, and the generated Stokes and anti-Stokes orders were captured using a charged-coupled device (Spiricon, SP503U) having a resolution of 640×480 pixels. Immediately before the crystal, the pump and Stokes beams had an energy per pulse of ~3 mJ. The pulse duration was extrapolated to be ~400 fs using a Grenouille (UPM-8-20, 18fs—180fs). With a spot size of 2mm (CCD) at the lens and for the given parameters, the peak intensity was estimated to be about ~3×10^9 W/cm^2, which assumes a diffraction-limited beam. For larger beam modes or for higher-order sidebands, the power needed to be increased to at most ~4mJ.
To produce Raman sidebands, we employed time-delayed chirped pulses in the beam crossing setup. Figure 2(a) illustrates how two delayed chirped-pulses in our experiments result in a frequency difference capable of being tuned to the Raman transitions in a nonlinear medium. For linear chirped pulses, the phase content has a quadratic time dependence, so that the instantaneous frequency is given by $\omega = \omega_0 + bt$. The two up-chirped pulses shown in Fig. 2(a) are delayed by $t_d$ and result in a frequency difference of $\Delta \omega = bt_d$ for all times $t$.

In our experiments, the first folding mirrors in the compressor of the laser was slightly detuned by about $\sim 1$mm. The sidebands were then found by adjusting the time delay between the two pulses using the translation stage shown in Fig. 2(b). In this way, the detuning $\Delta \omega = bt_d$ was found experimentally.

For completeness, the OAM-algebra derived by us in [15] for the helical LG beam of order $|\ell| = 1$ is briefly presented. In all further discussions, the different Raman orders will be signified by SN for Stokes and ASN for anti-Stokes, where N denotes the order. The pump is denoted by P and the Stokes by S. In Fig. 2(b) the level diagram shows two real states and a number of virtual states. To find the frequency of the radiation in the AS1 order, the nonlinear polarization can be written as the product of plane wave solutions $e^{i\theta} e^{-i\omega_s t} e^{i\omega_p t}$. The resulting electric field in the first anti-Stokes order then has a frequency of $\omega_{AS1} = 2\omega_p - \omega_s$. Multiplying through by $\hbar$ motivates a photon description in which the first anti-Stokes order can be described by the absorption of a pump photon followed by the stimulated emission of a Stokes photon with the subsequent absorption of another pump photon. The helical LG beams carry a phase factor of $e^{i\theta}$ in which its argument $\ell \theta$ comes along with the frequency, and therefore the same algebra $\ell_{AS1} = 2\ell_p - \ell_s$ follows. The amount of OAM in the AS2 order can be determined by the absorption of a pump photon followed by emission of a Stokes
photon with the subsequent absorption of an AS1 photon, so that 
\[ \ell_{AS1} = \ell_{P} - \ell_{S} + \ell_{AS1} = 3\ell_{P} - 2\ell_{S} \]. The absorption of the AS1 photon in the generation of the AS2 radiation demonstrates why this is a cascade process.

Continuing with this photon description, the OAM-algebra for anti-Stokes orders is found to be 
\[ \ell_{AS} = (n+1)\ell_{P} - n\ell_{S} \]. Using conservation of momentum \( \ell_{P} + \ell_{S} = \ell_{AS} + \ell_{S} \) the algebra for the Stokes orders is found to be 
\[ \ell_{S} = (n+1)\ell_{S} - n\ell_{P} \], which can be deduced by reversing the direction of the pump and Stokes arrows in Fig. 2(b). The image [bottom inset in Fig. 2(b)] shows measured sidebands up to the 19th anti-Stokes order. Due to spatially-dependent nonlinearities and dispersion, the orders do not lie along a line. The distortion can be manipulated by varying the overlap between the two beams.

3. Sideband generation with helical LG and IG beams

To check the functionality of our modified setup and to compare with results from our previous setup, on-axis computer generated holograms of helical Laguerre Gaussian beams [insets in Fig. 1(a)] were displayed on the SLM positioned in the four-port Michelson. The produced optical beam modes were then sent into the beam crossing setup and allowed to interact in the Raman-active crystal. When the device is used to produce off-axis holograms, amplitude information can also be encoded in the computer generated holograms by redirecting radiation between orders [2, 14]. This phase-amplitude encoding produces high fidelity beam modes but at the expense of pulse energy. In our setup, reduction of pulse energy from the three beam splitters, the mode-improving iris, chirp, and the reduction due to the efficiency of operating the SLM in off-axis mode resulted in a focal intensity insufficient to reliably produce Raman sidebands. For this reason, the SLM was operated in on-axis mode, which converts a large proportion of its input radiation into the desired mode.

In our previous setup, when radiation with \( \ell = \pm 1 \) was passed through the setup with mirror M3 in place, the topological charges in the pump and Stokes beams were found to have the same magnitude but were opposite in sign. In contrast, when mirror M3 was removed from the setup, the topological charges had the same magnitude and sign. This property of the beam carries over to higher-order modes with \( \ell > 1 \) and can be understood by a direct comparison with circularly polarized light. To show this, we decompose the Laguerre Gaussian beams with angular mode numbers \( \ell \) and radial mode number \( \rho = 0 \) in the Hermite Gaussian basis,
The derivation of Eq. (2) is given in Appendix A. Here $\text{sgn}(\ell)$ is the sign of the angular mode number $\ell$, $H_n$ are the Hermite polynomials, $\xi = \sqrt{2} x / w$ and $\eta = \sqrt{2} y / w$. Equation (2) represents the beam before reflection. Upon reflection the electric field experiences a phase shift of $\pi$ due to the larger refractive index of the mirror. Since linearly polarized light is used, this phase shift is unimportant. After reflection, we can perform a rotation to obtain the amplitude of the reflected beam. The $x$-axis is chosen to be the axis of rotation, so both $z$ and $y$ components will undergo a reflection,

$$H_{\ell+1}^*(-\eta) = (-1)^\ell H_{\ell}(-\eta).$$

(3)

In Eq. (3) the symmetry property $H_n(-x) = (-1)^n H_n(x)$ of the Hermite polynomial was used. Using Eq. (3), the reflected beam $\text{`LG}_{0,\ell}$ (denoted by a prime) can be found from the incident $\text{LG}_{0,\ell}$ beam in Eq. (2),

$$\text{`LG}_{0,\ell} = N_{\ell} \frac{w^2}{2} \sum_{\ell=0}^{\infty} \frac{|\ell|!}{k! (|\ell|-k)!} (i \text{sgn}(\ell))^k H_{|\ell|-k}^*(\xi) H_{\ell}(-\eta) e^{ikz}.$$  

(4)

In the reflected beam Eq. (4), a factor of $(-1)^\ell$ now appears. This factor can be combined with $\text{sgn}(\ell)$ to give $\text{sgn}(\ell)(-1) = \text{sgn}(-\ell)$. Comparing Eqs. (2) and (4), the sign of the mode

Fig. 3. Generation of Raman sidebands with LG beams having the same helicity $\ell_p = \ell_\xi$.
Columns 1—5: S1, S, P, AS1 and AS2; rows 1 and 3 are sidebands generated with $\ell = 1$ and 2 respectively. Rows 2 and 4 are the interferograms of those beams in rows 1 and 3 with the reference beam from the 4-port Michelson. The measured topological charges are consistent with that predicted by the OAM-algebra. Columns 6—9: S1, S, P, and AS1; row 1 are sidebands produced with $\ell = 3$ and row 2 shows the interferograms. Panels (a) and (b) show the intensity distribution and the interferogram of the AS1 order beam produced with $\ell = 5$. As can be seen, the multifurcation has broken up into 5 single bifurcations [denoted by the red dots in panel (b)].
number $\ell$ of the reflected beam is opposite to that of the incident beam $\ell_{LG_{0,\ell}} = \ell_{LG_{0,-\ell}}$. The OAM-algebra dictates that the amount of orbital angular momentum in the sidebands depends on the relative sign of $\ell_p$ and $\ell_S$, and in light of the results of Eq. (4) motivates the use of mirror $M_5$ in the setup that is needed for the correct interpretation of the sideband data generated with helical beam modes.

Figure 3 shows the beam profiles and interferograms of the generated sidebands using higher-order LG beams ($\ell = 1, 2, 3$) with mirror $M_5$ in place (balanced arms). For balanced arms, the mode number of the pump and Stokes are equal $\ell_p = \ell_S = \ell$. The OAM-algebra for this configuration reduces to $\ell_{AS} = \ell_n = \ell$. The topological charges in the pump and Stokes beams can be verified from the interferograms in Rows 2, 4 columns 1—5, and row 2 columns 6—9. In these images it can be seen that the multifurcations and bifurcations of all orders are facing in the same direction, and this indicates that $\ell_p = \ell_S = \ell$ verifying that the OAM-algebra is indeed satisfied.

![Beam profiles and interferograms](image)

Fig. 4. Generation of Raman sidebands with LG beam of the same order but opposite helicity $\ell_p = -\ell_S$ (unbalanced arms). Columns 1—5: S1, S, P, AS1 and AS2; rows 1 and 2, and rows 3 and 4 show sidebands generated with $\ell = 1$ and $\ell = 2$ respectively. Rows 2 and 4 are the interference of those beams in rows 1 and 3 with the reference beam for the Michelson. The measured topological charges are consistent with those predicted by the OAM-algebra $\ell_{AS} = (n+1)\ell_s - n\ell_p$. Column (a) shows images of the AS1 beams generated with $\ell = 4, 5, 6$ and 7 (rows 1—d respectively); and column (b) shows the interferograms. In all data, the topological charges were determined by counting multifurcation and fringes.

With mirror $M_5$ removed from the setup, the arms are unbalanced and the relative signs of $\ell$ between the pump and Stokes and the Stokes and anti-Stokes orders are opposite $\ell_{AS} = -\ell_{AS} = \ell$, so that the OAM-algebra reduces to $\ell_{AS} = (2n+1)\ell$ and $\ell_{AS} = -(2n+1)\ell$. Figure 4 shows sidebands produced in this manner. In this case, interferograms of the pump and Stokes beams show multifurcation facing each other showing that indeed $\ell_{AS} = -\ell_{AS} = \ell$.

The OAM-algebra for each order was verified by fringe counting. In our experiments, it was noted that the beam profiles in many of the sidebands were distorted, and the interferograms showed that the multifurcations in the higher-order modes separated into single bifurcations. This instability has been previously investigated and is found to commonly occur for high-order LG modes [19]. Despite the distortions, verification of the OAM in the generated sidebands is possible by identifying and summing the number of bifurcations or counting the fringes around a cluster of multifurcations. All data was found to be consistent with the OAM-algebra derived in [15]. We found that the quality of the generated sidebands is
sensitive to many experimental parameters. Such parameters include intensity, input beam size and chirp. When producing high order modes larger beam sizes result, and consequently the peak intensity drops. Further increase in intensity also results in distortion of the generated sidebands. Significant distortion and the presence of radiation from four-wave mixing were difficult to mitigate.

A natural extension of the OAM-algebra can be made to the OAM-carrying helical Ince-Gaussian beams Eq. (2). Like the helical-LG beams, the IG beams possess OAM. When the ellipticity parameter Eq. (1c) of the helical-IG beams is zero $\varepsilon = 0$, the helical-IG beams are equivalent to the helical-LG beams. This equivalence is a consequence of the relationship between cylindrical elliptic coordinates and cylindrical polar coordinates. When the ellipticity parameter is greater than zero $\varepsilon > 0$, the multifurcation of the beam splits into a string of bifurcations along a line joining the two foci of the coordinate. The total topological charge of the beam is then just equal to the sum of the bifurcations. For this reason, helical-IG beams of order $\ell > 1$ and $\varepsilon \geq 0$ were investigated.

Figure 5 shows generated sidebands of IG beams with $\ell = 2, 3, 4$ and 5 with varying non-zero ellipticities. As with the helical-LG beams, we observed distortion of the generated sidebands, but by inspection of the interferograms, the transfer of OAM into the sidebands was found to follow the same OAM-algebra as that for the helical-LG beams. For the current experimental conditions, an interesting effect was observed when the ellipticity parameter was varied between $\varepsilon = 0.1-0.4$. When $\varepsilon = 0$, all sidebands demonstrated a distortion in such a way that the bifurcations were separated about a line at ~70 degrees from the horizontal. This is similar to the instability of the higher-order LG beams. By varying $\varepsilon$, the beams were observed to take on a more symmetric shape (columns 2 and 7 in Fig. 5) and the interferograms showed a tendency for the individual bifurcations to group more towards the
center of the beams. This effect may present a method to actively correct for distortions in high-order OAM-containing beams in the focus or when delivered to a target.

4. Even Laguerre Gaussian modes

As shown above and in our previous work [15], a straightforward derivation based on parametric four-wave mixing allowed us to determine the orbital angular momentum algebra of the generated sidebands for the helical-LG beams. We now turn our attention to the even and odd Laguerre Gaussian beams of Eq. (1). The solutions of these beams are similar to those of the helical-LG beams with the exception that the total OAM sums to zero. Unlike the OAM-algebra for the helical-LG beams, the derivations for the even and odd beams are more involved. Our considerations in this section will be for the even LG beams; however the results directly carry over to the odd LG beams by adding \( \pi/2 \) to the arguments of the cosine functions. To gain understanding of how OAM from the even LG beams affect the generation process, we derive the OAM-algebra with the help of four-wave mixing relations and by using the energy level diagram in Fig. 2(b) as a guide. A consequence of this scheme is the neglecting of terms in the polarizability that do not satisfy the cascaded Raman process.

In nonlinear optics, the polarizability can generally be expanded as a power series in the electric field \( P(t) = \varepsilon_0 \chi^{(1)} E(t) + \varepsilon_0 \chi^{(2)} E^2(t) + \varepsilon_0 \chi^{(3)} E^3(t) + \cdots \), where the \( \chi^{(n)} \) are the nonlinear susceptibilities [20]. The scalar wave equation for the generated radiation field is

\[
\nabla^2 E - \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P_{NL}}{\partial t^2}.
\]

Since we are interested in the OAM content of the beams, we assume that the amplitudes of these beams are uniform in the radial direction, and that the changes in the amplitudes upon propagation are negligible. The nonlinear polarization will then be a function of the angular coordinate only. With these considerations, we take the field of the ASN order to have the general form \( E = E_{\text{ASN}}(\theta) e^{i(kz-\omega t)} \). Substituting this field into Eq. (5) yields

\[
\frac{1}{r^2} \frac{\partial}{\partial \theta} E_{\text{ASN}}(\theta) e^{i(kz-\omega t)} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P_{NL}}{\partial t^2}.
\]

For the even LG beams, the nonlinear polarization \( P_{NL} \) can be found by taking the field of the pump beam to be \( E_p(\theta) = \cos(\ell_\varphi \theta) e^{i(k_p z - \omega_p t)} \) and that of the Stokes beam to be \( E_s(\theta) = \cos(\ell_\varphi \theta) e^{i(k_s z - \omega_s t)} \). Following the energy level diagram in Fig. 2(b), the field of the AS1 beam is found to be \( P_{\text{AS1}} = E_p E_s^* E_p \). For the \( N \)th sideband, the nonlinear polarization is \( P_{\text{ASN}} = E_p^{N+1} E_s^N \) or written out explicitly,

\[
P_{NL} \propto \cos^{N+1}(\ell_\varphi \theta) \cos^N(\ell_\varphi \theta) e^{i((N+1)k_p - Nk_s)z - i(N+1)\omega_p - N\omega_s) t}.
\]

The nonlinear polarization in Eq. (7) contains only terms that satisfy the cascaded Raman process. By substituting this polarization into Eq. (6), one can immediately determine the frequency and wave number of the radiation in the generated sidebands,

\[
\frac{1}{r^2} \frac{\partial^2 E_{\text{ASN}}(\theta)}{\partial \theta^2} e^{i(kz-\omega t)} = -\frac{1}{\varepsilon_0 c^2} \left[ ((N+1)\omega_p - N\omega_s) \cos^{N+1}(\ell_\varphi \theta) \cos^N(\ell_\varphi \theta) e^{i((N+1)k_p - Nk_s)z - i((N+1)\omega_p - N\omega_s) t} \right].
\]
By comparing the frequency and wavenumbers on both sides of Eq. (8), it is found that $k_{\text{ASN}} = (N+1)k_p - NK_s$ and $\omega_{\text{ASN}} = (N+1)\omega_p - N\omega_s$. The first of these equations leads to phase matching (momentum conservation) and the second leads to the frequency of each sideband (energy conservation).

To obtain an analytical expression, we note that the right hand side of Eq. (8) is periodic in $\theta$ and motivates a periodic solution of the form $E_{\text{ASN}}(\theta) = \sum a_n \cos(n\theta)$. With this substitution in Eq. (8) and with $B = [(N+1)\omega_p - N\omega_s] / c^2$, Eq. (8) can then be written as

$$-\frac{1}{r^2} \sum a_n^2 \cos(n\theta) = -B \cos^{N+1}(\ell_p \theta) \cos^N(\ell_s \theta). \quad (9)$$

The summation on the left hand side can be found by repeated use of $2 \cos^2(\ell_p \theta) = 1 + \cos(2\ell_p \theta)$ and $2 \cos(\ell_p \theta) \cos(2\ell_p \theta) = \cos[(2\ell_p + \ell_s)\theta] + \cos[(2\ell_p - \ell_s)\theta]$ on the product of cosines on the right hand side of Eq. (9). Using these identities, the product can be separated into the sum of individual cosine terms. This tedious exercise leads to the following expansion,

$$E_{\text{ASN}}(\theta) = \sum_{k=0}^{N} \sum_{j=-N}^{N} a_{jk} \cos[(k \ell_p + j \ell_s)\theta] + (1 + (-1)^{N+1}) \frac{1}{2} \sum_{j=-N}^{N} a_{j0} \cos(j \ell_s \theta) \quad (10)$$

Equation (10) is the main result for describing the OAM-algebra of the even LG beams. The arguments of the trigonometric terms provide the OAM content of the generated sidebands. The sums in Eq. (10) are taken over even values when the upper bounds are even, and odd when the upper bounds are odd. This is the OAM-algebra generalized to the even LG beams.

As an example, when $N=1$, we expect to find indications of even LG beams of $LG^{0,\ell}_{0,\ell}$, $LG^{0,2\ell}_{0,\ell}$ and $LG^{0,2\ell}_{0,\ell}$ in the AS1 radiation. In the special case when the magnitudes of $\ell$ for the pump and Stokes are the same, but the sign is allowed to be positive or negative, we respectively find

$$E_{\text{ASN}}(\theta) = \sum_{k=0}^{N} \sum_{j=-N}^{N} a_{jk} \cos[(k + j) \theta] + (1 + (-1)^{N+1}) \frac{1}{2} \sum_{j=-N}^{N} a_{j0} \cos(j \theta), \quad (11a)$$

$$E_{\text{ASN}}(\theta) = \sum_{k=0}^{N} \sum_{j=-N}^{N} a_{jk} \cos[(k - j) \theta] + (1 + (-1)^{N+1}) \frac{1}{2} \sum_{j=-N}^{N} a_{j0} \cos(j \theta) \quad (11b)$$

Since the sums over $j$ in Eqs. (11a) and (11b) run from $-N$ to $N$, both equations are the same. Equations (11) indicate that generating sidebands with mirror M5 in place or removed will not change the observed radiation pattern. This is in contrast to that found for both the helical LG and the LG beams.

Experimental results of the generation of Raman sidebands with the pump and Stokes beams in the modes $LG^{0}_{\ell,0}$ and $LG^{0}_{\ell,0}$ are shown in Fig. 6. Row 1 shows the S1, S, P, AS1 and AS2 beams (columns 1—5 respectively) for the $LG^{0}_{\ell,0}$ modes, and row 2 shows the same orders for the $LG^{0}_{\ell,0}$ beam. From Eq. (10), the OAM-algebra for the S1 and AS1 orders ( $N=1$ ) suggests that the resulting beams will have contributions from $\ell, -3\ell$ and $3\ell$. For the $LG^{0}_{\ell,0}$ modes ($\ell = 2$) in row 1, the expected contributions are from $LG^{0}_{\ell,2}$ and $LG^{0}_{\ell,6}$, and for the $LG^{0}_{\ell,0}$ modes in row 2 the expected contributions are from $LG^{0}_{\ell,0}$ and $LG^{0}_{\ell,0}$. For
the $\ell = 2$ modes in the top row, the $LG_{e,2}^\ell$ contribution is the same as the pump and Stokes and has 4

![Image](image_url)

Fig. 6. Generation of Raman sidebands with $\ell$LG beams. Columns a—c: S1, S, P, AS1 and AS2; row 1 are sidebands generated with $\ell=2$ and Row 2 are sidebands generated with $\ell=3$. For the even (odd) LG beams, the number of angular nodes is $2\ell$. From the OAM-algebra (see text, Eq. (10)), the AS1 and S1 orders are expected to have modal contributions from $\ell$ and $3\ell$. So for $LG_{e,2}^\ell$ in the first row, the S1 and AS1 order are expected to have similarities with $LG_{e,2}^\ell$ and $LG_{e,6}^\ell$ modes having 4 and 12 nodes. In the bottom row $\ell=3$ the S1 and AS1 orders are expected to have contributions from the $LG_{e,3,3}^\ell$ and $LG_{e,9}^\ell$ modes. These modes have 6 and 18 nodes respectively. For AS2 sideband, the OAM-algebra results in angular mode numbers of $\ell$, $3\ell$ and $5\ell$. For $LG_{e,3}^\ell$ (top row) the AS2 order is expected to have contributions from $LG_{e,2}^\ell$, $LG_{e,8}^\ell$ and $LG_{e,10}^\ell$ (4, 12 and 20 angular nodes) and for $LG_{e,5}^\ell$ (bottom row) the AS2 order is expected to have contributions from $LG_{e,1}^\ell$, $LG_{e,9}^\ell$ and $LG_{e,15}^\ell$ (6, 18 and 30 angular nodes).

nodes, while the contribution from the $LG_{e,0,6}^\ell$ mode has 12 nodes. This result is consistent with the measured S1 and AS1 orders in row 1. For the S1 and AS1 orders in row 2, we expect contributions from $LG_{e,0,3}^\ell$ which are the pump and the Stokes modes and from $LG_{e,0,9}^\ell$ which has 18 angular nodes. These results are in excellent agreement with the measured data. For the AS2 order, Eq. (10) predicts that this order is expected to have contribution from even LG beam of mode orders $LG_{e,1}^\ell$, $LG_{e,0,9}^\ell$ and $LG_{e,0,15}^\ell$, which have angular nodes of 6, 18 and 30. Again these results are consistent with the measured data and suggest that Eq. (10) is capable of describing the angular momentum content of the generated sidebands.

Because the holographic generation of the pump and Stokes beams is an imperfect process, it may happen that impurities in the generated modes produce similar modal lobes in the generated Raman orders. Although inspection of the imaging suggests that this is not a likely conclusion, nevertheless we decided to investigate the generation process further by blocking a modal lobe of the Stokes beam. Figure 7 shows the result of this experiment. Row 1 shows the S, P, and AS1 orders (from left to right) with an even $LG_{e,2}^\ell$ beam. In this case, no modal lobes were blocked. The data in row 2 shows the measured result when a single modal lobe of the Stokes beam is blocked. In this case the generated AS1 order shows a nearly identical pattern as compared to the case with no blocking. This suggests that the lowest order modal contribution predicted by the OAM algebra is being generated in the nonlinear process. The result also indicates that the entire mode structure is coherently
participating in the generation process, and that the highly nonlinear nature of the process is not causing each modal lobe to act independently. Finally, the Hermite Gaussian modes can be expanded in the even and odd LG basis by a transformation. For completeness, the expressions for the expansions are given in Appendix B. For the work presented here, it can be seen that the first row in Fig. 6 and the modes in Fig. 7 are both an \( \text{LG}_{0,2}^{\circ} \), and a \( \text{HG}_{1,1} \) rotated at a 45 degree angle and the expansion is given in Eq. (26).

\[
\text{Fig. 7. Generation of Raman sidebands with even } \text{LG} \text{ beams having a blocked lobe. Columns 1—3: S, P, and AS1. Row 1 is generated with } \ell = 2 \text{. Row 2 is generation with even } \ell \text{ LG beams having } \ell = 2 \text{. In the last row, a modal lobe was blocked to investigate the generation of the lowest order contribution to the OAM-algebra which in this case is } \text{LG}_{0,2}^{\circ} \text{. The AS1 orders for both scenarios (blocked and unblocked) are nearly identical demonstrating the generation of the predicted modes and that the entire modes structures of the pump and Stokes are participating in the generation process.}
\]

5. Discussion and summary

In the synthesis of few-cycle pulses of radiation from the combination of Raman sidebands, it is possible to produce a short pulse in a pure transversal mode when using the helical LG and IG beams. This is because when \( \ell_s = \ell_p \), the OAM-algebra shows that the OAM in each order is the same. The question as to whether a similar situation can occur for the even (odd) LG and the HG beams naturally arises. The extended algebra of Eq. (10) shows that all the sidebands will have contribution from many modes, but they will all have a contribution from the generating mode. It may therefore be possible to spatially filter out the mode impurities before combining the sidebands in the synthesis of few-cycle pulse.

In conclusion, we have generated Raman sidebands using various beam modes and investigated the nonlinear interaction of these beams in a Raman active crystal. We have generalized the OAM-algebra to include the OAM content of higher-order LG and IG beams, even and odd LG beams, and HG beams. Measurements of interference patterns produced with the reference beam in a simultaneous Young double slit experiment provided quantitative confirmation of the derived results.

Appendix A: Decomposition of the \( \text{LG}_{n,l} \) beams in the Hermite Gaussian basis

The scalar equations of the helical-LG and Hermite Gaussian beams at the waist \( z = 0 \) is given by
\[ \text{LG}_{0,0} = N_0^0 \left( \frac{\sqrt{2r}}{w} \right)^{|l|} e^{il\theta} e^{-r^2/w^2} \]  

(12)

\[ \text{HG}_{mn} = N_{mn} H_n \left( \frac{\sqrt{2x}}{w} \right) e^{-x^2/w^2} H_m \left( \frac{\sqrt{2y}}{w} \right) e^{-y^2/w^2} \]  

(13)

Where \( N_0^0 \) and \( N_{mn} \) are normalization constants. For simplicity we make the substitution \( \xi = x\sqrt{2} / w \) and \( \eta = y\sqrt{2} / w \), where \( r^2 = \xi^2 + \eta^2 \). The relationship between the two basis sets can be found by the expansion,

\[ \text{LG}_{0,0} = \sum_{m,n} a_{mn} \text{HG}_{m,n} \]  

(14)

The coefficients \( a_{mn} \) can be found by multiplying both sides of Eq. (14) by \( \text{HG}_{m,n} \) and integrating. This is known as Fourier’s trick. For normalized functions, the right hand side gives \( a_{mn} \). The left hand side gives the integral

\[ I_{LHS} = N_0^0 N_{mn} \int \left( \frac{\sqrt{2r}}{w} \right)^{|l|} e^{il\theta} H_n(\xi) H_m(\eta) e^{-r^2/w^2} dxdy. \]  

(15)

The integral in Eq. (15) can be recast into an integral with Cartesian coordinates using the binomial theorem on the phase factor,

\[ e^{il\theta} = \sum_{i=0}^{|l|} \left[ i \text{sgn}(i) \sin(\theta) \right]^i \cos(\theta)^{|l|}. \]  

(16)

Substituting Eq. (16) into Eq. (15) and using \( x = r \cos(\theta) \) and \( y = r \sin(\theta) \) we get

\[ I_{LHS} = N_0^0 N_{mn} \sum_{i=0}^{|l|} \left[ i \right] \left( \text{sgn}(i) \right)^i \int \xi^{|l|+i} \eta^i H_n(\xi) H_m(\eta) e^{-r^2/w^2} dxdy. \]  

(17)

The integral can be split into two separate integrals: one over \( \xi \) and the other over \( \eta \). These integrals with the help of Rodrigues formula can be rewritten as,

\[ \int \eta^k e^{-\eta} H_m(\eta) d\eta = (-1)^m \int \eta^k e^{-\eta} \frac{d^m}{d\eta^m} (e^{-\eta}) d\eta, \]  

(18)

\[ \int \xi^{|l|+i} H_n(\xi) e^{-\xi} d\xi = (-1)^i \int \xi^{|l|+i} e^{-\xi} \frac{d^i}{d\xi^i} (e^{-\xi}) d\xi. \]  

(19)

The first integral is non-zero only when \( m = k \) and the second integral is nonzero only when \( |l| - k = n \) giving the results

\[ \int \eta^k e^{-\eta} H_m(\eta) d\eta = (-1)^k k! \sqrt{\pi}, \]  

(20)

\[ \int \xi^{|l|+i} H_n(\xi) e^{-\xi} d\xi = (-1)^i |l|! \sqrt{\pi}. \]  

(21)

With a little algebra and using Eqs. (17), (20) and (21), the expansion Eq. (14) can be written as
\[ \text{LG}_{n_1,n_2} = N_0 \frac{w^2}{2} \left( \frac{-1}{2 \Gamma} \right) \sum_{k=0}^{[|n_1|-|n_2|]} \frac{|k|!}{k!(|n_1|-|n_2|)!} (\text{sgn}(t))^k H_{|n_1|+|n_2|} (\xi) H_{|n_1|} (\eta) e^{2k}. \]  

(22)

**Appendix B: Decomposition of the \( \text{HG}_{nm} \) beams in the Laguerre Gaussian basis**

The expansion of the Hermite Gaussian beam in the Laguerre Gaussian basis set has been previously calculated [21]. Using this expansion and the OAM-algebra found from Eq. 12, the OAM content of a Hermite Gaussian mode may be determined. There are four equation for the expansion depending on the parity of the mode indices such as even-even, odd-even, even-odd, and odd-odd. When both indices are even, the expansion is

\[
\begin{align*}
\text{HG}_{2k,2j} &= \frac{(-1)^j}{2^{j+k+1}} \sum_{q=0}^{j+k} q!(2j+k+1-q)! \text{LG}_{j+k,0}^q \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k,0}^q \right] \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k,0}^q \right].
\end{align*}
\]

(23)

and for the odd-even case

\[
\begin{align*}
\text{HG}_{2k+1,2j} &= \frac{(-1)^j}{2^{j+k+1}} \sum_{q=0}^{j+k} q!(2j+k+1-q)! \text{LG}_{j+k+1,1}^q \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k+1,1}^q \right] \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k+1,1}^q \right].
\end{align*}
\]

(24)

and for the even-odd case

\[
\begin{align*}
\text{HG}_{2k,2j+1} &= \frac{(-1)^j}{2^{j+k+1}} \sum_{q=0}^{j+k} q!(2j+k+1-q)! \text{LG}_{j+k,1}^q \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k,1}^q \right] \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k,1}^q \right].
\end{align*}
\]

(25)

and lastly for the even-even

\[
\begin{align*}
\text{HG}_{2k+1,2j+1} &= \frac{(-1)^j}{2^{j+k+1}} \sum_{q=0}^{j+k} q!(2j+k+1-q)! \text{LG}_{j+k+1,0}^q \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k+1,0}^q \right] \\
&= \left[ \sum_{q=0}^{j+k} (-1)^q \binom{2j+k+1-q}{j+1} \frac{1}{2^{j+k+1}} \text{LG}_{j+k+1,0}^q \right].
\end{align*}
\]

(26)

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