Radial modal transition of Laguerre-Gauss modes in second-harmonic generation

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Optical orbital angular momentum conservation and their corresponding selection rules have been exhaustively studied for various nonlinear optical processes; however, the radial mode has not been systematically studied. In this work, we revisit second-harmonic generation (SHG) pumped by Laguerre-Gaussian modes with various radial structures and study, both theoretically and experimentally, the transition of the radial modal of the SHG fields from the origin to the far field in detail. We provide a general theory for describing the radial structure of the SHG field and verify its accuracy via experiments. Our results show that there is no concise selection rule for the radial index because of the unstable radial structure of the SHG field. However, for some specific cases, a concise selection rule for the far-field radial structure can still be determined.

Soon after Allen et al. reported on optical orbital angular momentum (OAM) [1], research on OAM in nonlinear optics began with the second-harmonic generation (SHG) of Laguerre-Gaussian (LG) modes (paraxial carrier of OAM) [2]. The azimuthal modal transition (or OAM selection rule) determined from their work, i.e., SHG fields carry twice the OAM of the pump, provided straightforward insights into OAM conservation for nonlinear interactions in the photon picture. Thereafter, OAM conservation and the corresponding selection rules in various nonlinear optical processes were studied in depth, such as sum-frequency generation (SFG), four-wave mixing, high-order-harmonic generation, and light–sound (or other matter waves) interactions [3–13]. Today nonlinear interactions involving OAM carrying light have become a crucial method to shape structured light and manipulate high-dimensional photons [14–16], and these interactions are of interest for both practical applications and fundamental optics. Recently, research on the LG modes has gradually begun to focus on their radial modes, in particular, for their potential application in quantum information [17–24]. For quantum application, the radial modal transition for nonlinear interactions plays a vital role in quantum memory and frequency transducers [25–29], however, they have yet to be studied quantitatively [30–33]. To address this issue, we revisit the SHG of LG modes with radial structures and study the radial modal transition of the second-harmonic field upon paraxial propagation.

The free space wavefunction of LG modes in cylindrical coordinates can be fully determined by the wave vector $k$, origin beam waist $w_0$, and two spatial (i.e., azimuthal and radial) indices $\ell$ and $p$, given by [1]:

$$LG_p^\ell(r,\varphi, z) = G(r, z; k, w_0, p, \ell)E_\ell^\ell(\gamma)\exp(-i\varphi),$$

where $G(r, z; k, w_0, p, \ell)$ denotes a Gaussian-type transverse amplitude, $E_\ell^\ell(\gamma)$ is the Laguerre polynomial with the mode orders $p$ and $|\ell|$, and the variable $\gamma = 2r^2/w_0^2[1 + (z/z_R)^2]$ (here $z_R = kw_0^2/2$ is the Rayleigh length). It should be noted that $E_\ell^\ell(\gamma)$ shapes the transverse amplitude in the radial dimension, i.e., the radial index is directly related to the Laguerre polynomial. To be more specific, the zero solution of $E_\ell^\ell(\gamma)$ specifies the number ($N = p \geq 0$) and position of antiphase dislocations along the radial coordinate, giving a specific multiring structure.

Among the two spatial indices, the azimuthal index $\ell$ (also referred to as topological charge) can be regarded as the eigenvalue of the OAM operator with respect to the $z$-axis $\hat{L}_z = -i\hbar(\partial/\partial\varphi)$, i.e., $\hat{L}_zLG_p^\ell(r,\varphi, z) = i\ell LG_p^\ell(r,\varphi, z)$ because of the twisted phase profile $\exp(i\varphi)$ within the LG modes. Here, both the operator $\hat{L}_z$ and the associated observable are independent to the other beam parameters, so the azimuthal quantum number is a conserved quantity corresponding to OAM conservation in paraxial propagation. Based on this well-defined conserved parameter, the selection rules of $\ell$ in various nonlinear interactions can be inferred. For the radial index $p$, its operator can indeed be defined via a relation $\hat{P}LG_p^\ell(r,\varphi, z) = pLG_p^\ell(r,\varphi, z)$, however, this operator, unlike $\hat{L}_z$, is correlated with other observables including $w_0$, $\ell$, and even $z$ [19]. That is, the radial quantum number is not a conserved quantity of motion and can only be well defined on knowing all the relevant beam parameters. For this reason, most recent experiments exploring the quantum properties of the radial quantum number have adopted imaging systems to match the relevant beam parameters between the sender and receiver [20–22]. Because of this complexity, determining the selection rule of the radial index in a nonlinear interaction is not straightforward, and in this work, we start from the simplest nonlinear interaction SHG (a particular case of SFG).
For simplicity and without loss of generality, we assume that SHG is driven by a quadratic beating of two monochromatic and collimated LG modes with the same $k$ and $w_0$, but different spatial indices. In Appendix A, we also provide a more general theory describing SFG between two modes with different frequencies and beam waists. As the source of the second-harmonic field, the excited nonlinear polarization (NP) at the $z_0$ plane can be expressed as

$$\mathbf{P}^{\text{2sh}} = kE^{\text{2sh}}(r, \varphi, z_0),$$

where $k = e_2(n_2/c)$ denotes the nonlinear coupling coefficient and $E^{\text{2sh}}(r, \varphi, z_0) = LG^0_{p1}(r, \varphi, z_0) + LG^1_{p1}(r, \varphi, z_0)$ is the quadratic beating field. From this NP, we can infer that the SHG undergoes doubling transitions for both the momentum (or frequency) and OAM, corresponding to $\exp[i(k\omega_0z)] \rightarrow \exp[i(2k\omega_0z)]$ and $\exp[i(2\varphi)] \rightarrow \exp[2i(\varphi)]$, respectively. While, for the radial structure, at this stage we can only conclude that the radial structure of the amplitude at the $z_0$ plane is governed by term $E^{\text{2sh}}(r, z_0) = L^0_{m0}(\cdot) \ast L^0_{m0}(\cdot)$ [see Appendix A for details], and the beam waist reduces to $w_0 (\sqrt{2})^{-1}$. Note, if $p_1$ and $p_2 = 0$, this second-harmonic mode is not an eigen solution of the paraxial wave equation, leading to an unstable amplitude structure during propagation.

Therefore, to reveal the complete transition rule of the radial structure, it is necessary to obtain the traveling wave equation of the second-harmonic field $E^{\text{2sh}}(r, \varphi, z)$. The general solution of the second-harmonic field can be derived using the Collins propagator with $E^{\text{2sh}}(r, \varphi, z_0)$ as the pupil function [34–38], which can be expressed as [see Appendix A for details]:

$$E^{\text{2sh}}(r, \varphi, z) = U(r, z; 2k, w_0, p_{1z}, \xi_{1z}) L^{\text{2sh}}(r, z) \exp(-i\varphi)$$

$$L^{\text{2sh}}(r, z) = \sum_{m=p}^{m=p} q_m(z) L^0_{m0}(\cdot),$$

where $U(r, z; 2k, w_0, p_{1z}, \xi_{1z})$ is the transverse amplitude, $E^{\text{2sh}}(r, z)$ is a series solution governing the radial structure, and $m = \xi_1 + \xi_2$ and $n = \left|\xi_1\right| + \left|\xi_2\right| - \left|\xi_1 + \xi_2\right|/2$. Using Eq. (2), the three-dimensional (intensity and phase) structure of the second-harmonic field can be fully predicted, or rather, the transverse structure from the origin plane $(z_0)$ to the far field $(z_\infty)$.

$$E^{\text{2sh}}(r, \varphi, z) = U(r, z; 2k, w_0, p_{1z}, \xi_{1z}) L^{\text{2sh}}(r, z) \exp(-i\varphi)$$

Equation (2) contains a twisted phase profile of the form $\exp(i\varphi)$, which means $E^{\text{2sh}}(r, \varphi, z)$ carrying a well-defined OAM of $m\hbar$ per photon. Thus, a concise selection rule of the azimuthal index for SHG can be determined, i.e., $m = \xi_1 + \xi_2$. While for the radial structure, we note that the governing term $E^{\text{2sh}}(r, z)$ consists of a series of Laguerre polynomials, where the weight coefficients $q_m(z)$ and the complex variable $\zeta_z$ are both functions of $z$. This indicates the radial structure of the second-harmonic field is unstable upon propagation. Moreover, according to the order of the polynomial, i.e., $n + j$, we know that the number of antiphase dislocations ($N$) at a given propagation plane depends on both the azimuthal and radial indices of the pump fields.

Equation (2) reveals that it is impossible to determine a concise selection rule for the radial structure in propagation. Nonetheless, at the far field Eq. (2) will turn into a Fourier transform of the $z_0$ plane, i.e., $E^{\text{2sh}}(r, \varphi, z_0) \rightarrow \mathcal{F}[E^{\text{2sh}}(r, \varphi, z_0)]$, at which, more remarkably, $E^{\text{2sh}}(r, z)$ becomes a real function (i.e., $\zeta_z \rightarrow \mathbb{R}$ at $z_\infty$) and is independent of $z$. Therefore, except for the origin plane, one can obtain a propagation independent selection rule of the far-field radial structure of the second-harmonic field via a Fourier transform [see Appendix A for details]:

$$\mathcal{F}[E^{\text{2sh}}(r, \varphi, z_0)] = U(r, z_0; w_0, p_{1z}, \xi_{1z}) L^{\text{2sh}}(r, z_0) \exp(-i\varphi).$$

Here, $L^{\text{2sh}}(r, z_0)$ is a series solution of the radial coordinate that governs the radial structure of the far-field amplitude. Remarkably, for the special case of $p_1 = p_2 = p$, the series solution can be further factorized into two concise forms:

$$L^{\text{2sh}}(r, z_0) = L^0_{p0}(\cdot) \ast L^0_{p0}(\cdot), \quad \text{for } \xi_1 \times \xi_2 > 0$$

$$L^{\text{2sh}}(r, z_0) = L^0_{p0}(\cdot) \ast L^0_{p0}(\cdot), \quad \text{for } \xi_1 \times \xi_2 < 0.$$
FIG. 1. Simulated radial structure of the second-harmonic fields from the origin plane to the far field, where (a) corresponds to the SHG of $LG_0 \star LG_0^\ast$, and (b)–(c) correspond to the SHG of $LG_0 \star LG_1^\ast$, $LG_0^\ast \star LG_1$, and $LG_0^\ast \star LG_1^\ast$, respectively.

Next, the above analysis was verified by comparing the simulations with their corresponding experimental results, where the beam profiles of the second-harmonic field from the $z_0$ to $z_\infty$ plane were chosen as the characteristic observable. Figure 1 shows the experimental setup, a type-II SHG with two orthogonally polarized LG modes was used as the experimental platform. A narrow linewidth 800 nm laser was first converted to a perfect horizontally polarized TEM$_{00}$ mode by passing through a spatial filter in combination with a polarizing beam splitter (PBS), which was then incident on a spatial light modulator (SLM). The Damman gratings based on complex amplitude modulation were used to simultaneously generate a pair of LG modes with the same $w_0$ but different spatial indices [40,41]. The two LG modes were converted to orthogonal polarizations via a half-wave plate (HWP) and then combined into a copropagating beam, i.e., a vector mode, using a 4f-imaging system with a polarizing grating (PG). To demonstrate SHG driven by collimated beams, the prepared ‘vector mode’ was then loosely focused into a 3-mm-long type-II PPKTP. After filtering the generated 400 nm light, another 4f-imaging system and CCD were employed to record the beam profiles from $z_0$ to $z_\infty$.

For the experiments, we first focus on case (i), Fig. 3(a) shows the observed beam profiles of the SHG field pumped by $LG_p \star LG_p^\ast$ and $LG_p^\ast \star LG_p$ with $p = 0, 1, \text{ and } 2$. We see that the observation shown in the left column exactly coincides with the theoretical reference shown in the right column. Moreover, as predicted above, although the radial structure of the SHG fields rapidly changed during propagation, for the case $\ell_1 \times \ell_2 > 0$, their amplitude profiles eventually went back to their original form at the far field ($z_\infty$ plane). Next, case (ii) is further considered, Fig. 3(b) shows the experimental observation and associated theoretical reference for the SHG of $LG_p \star LG_p^\ast$ and $LG_p^\ast \star LG_p^\ast$ with $p = 0, 1, \text{ and } 2$. The experimental observations again exactly match the theoretical predictions, with further comparison data provided in Appendix B. Compared with the $z_0$ plane, the results show that the radial antiphase dislocations of the
SHG field at the $z_\infty$ plane exist “azimuthal to radial modal conversion”. It is worth noting that the “azimuthal to radial modal conversion” also occurs for the SHG pumped by LG modes with $p=0$, i.e., SHG of \( LG_0^p \ast LG_0^{-1} \) and \( LG_0^p \ast LG_0^{-2} \). Here, it was observed that the outer ring of the beam profile at the far field predicted by our theory was much weaker than that recently reported in a relevant literature report [31], and our experimental observation better matched the theory. Finally, more general examples of the two cases, i.e., SHG of \( LG^1_1 \ast LG^2_1 \) and \( LG^{-1}_1 \ast LG^{-1}_1 \), were compared. Figure 3(c) shows the corresponding results that confirm the accuracy of the theory once again. Besides, it should be noted that, both for the simulation and experiments, the two pump beams were assumed to have the same $k$ and $w_0$ for simplicity. In Appendix A, we also provide a theory for a more general SFG driven by two beams with different $k$ and $w_0$.

FIG. 3. The comparison between the observed radial structure of the SHG and the associated theoretical references. (a) Results for \( LG^1_1 \ast LG^3_1 \) and \( LG^2_1 \ast LG^1_1 \) with $p = 0, 1, \text{and } 2$; (b) for \( LG^1_2 \ast LG^3_1 \) and \( LG^3_2 \ast LG^2_1 \) with $p = 0, 1, \text{and } 2$; and (c) for \( LG^1_1 \ast LG^1_1 \) and \( LG^1_1 \ast LG^1_1 \).
In summary, a detailed theoretical analysis and experimental demonstration of the radial modal transition of the LG modes in SHG (and SFG) was presented. Specifically, a general solution for describing the radial structure of the second-harmonic field pumped by LG modes upon propagation was provided, as well as a special solution for several specific cases in the far field. Our results show that the radial structure of an LG mode that underwent a nonlinear interaction is usually not stable on free-space propagation. Compared with the selection rule for the OAM, there was no concise selection rule for the radial index. However, for some specific cases, concise selection rules could be determined, as shown in Eq. (4) for the far-field radial structure. Finally, an autocorrelation relation present in Eq. (4) indicates that SHG (and SFG) with LG modes may provide direction for applications based on light-field correlation.

ACKNOWLEDGMENT

This work was supported by the National Natural Science Foundation of China (Grant Nos. 11934013, 61975047, and 11874102).

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Appendix A  Theoretical Frame

The wavefunction of Laguerre-Gaussian (LG) mode in cylindrical coordinates \((r, \varphi, z)\), used in simulations, is given by

\[
LG_p^l(r, \varphi, z) = \sqrt{\frac{2^{p+1}}{\pi (p+|l|)! w(z)}} \left(\frac{\sqrt{v}}{w_z}\right)^{|l|} \exp \left[-\frac{r^2}{w_z^2}\right] \times L_p^l \left(\frac{2r^2}{w_z^2}\right) \exp \left[-i \left(kz + \frac{kr^2}{2R_z} + \ell \varphi - i\phi_0\right)\right],
\]

where \(w_z = \sqrt{w_0^2 + (z/z_R)^2}\), \(R_z = z^2 + z_R^2\), and \(\phi_0 = (2p + |l| + 1)\arctan(z/z_R)\) denote the beam waist, radius curvature, and Gouy phase upon propagation (here \(z_R = k w_0^2 / 2\) is the Rayleigh length, respectively), and \(L_p^l(\gamma)\) is the Laguerre polynomial with the mode orders \(p\) and \(|l|\), given by \(L_p^l(\gamma) = \sum_k ((|l| + p)!! / (|l|!! k!! l!!)) \gamma^{k - |l|} / (k!!(p - k)!)\). In the main text Eq. (S1) is abbreviated as

\[
LG_p^l(r, \varphi, z) = G(r, z; k, w_0, p, l) L_p^l(\gamma) \exp(-i \varphi),
\]

corresponding to Eq. (1).

For a SHG (or SFG) driven by \(LG_{p_1}^l(r, \varphi_0)\) and \(LG_{p_2}^l(r, \varphi_0)\), the wave source of second-harmonic fields, i.e., nonlinear polarization, is excited by the quadratic beating field, given by

\[
E^{2\omega}(r_0, \varphi_0) = LG_{p_1}^l(r_0, \varphi_0) \times LG_{p_2}^l(r_0, \varphi_0)
\]

\[
= \frac{2}{\pi} \left(\frac{p_1 p_2!}{(|l_1| + p_1)!(|l_2| + p_2)!}\right) \left(\frac{\sqrt{v}}{w_1 w_2}\right)^{|l_1 + l_2|} \exp \left[-r \left(\frac{1}{w_1^2} + \frac{1}{w_2^2}\right)\right] \exp[-i (l_1 + l_2)] L_{p_1}^{l_1} \left(\frac{2r^2}{w_1^2}\right) L_{p_2}^{l_2} \left(\frac{2r^2}{w_2^2}\right),
\]

where \(w_1\) and \(w_2\) denote the beam waists of two pumps. Therefore, the traveling wave equation of the second-harmonic field \(E^{2\omega}(r, \varphi, z)\) can be derived by using Collins propagator with \(E^{2\omega}(r_0, \varphi_0)\) as the pupil function, which is given by

\[
E^{2\omega}(r, \varphi, z) = \frac{i}{\lambda z} \exp(-ikz) \int d\varphi_0 \mathcal{F} \exp \left[-\frac{ik}{2z} [r_0^2 - 2r_0 \cos(\varphi - \varphi_0) + r^2]\right]
\]

\[
= \frac{1}{\lambda z} \sqrt{\frac{2(|l_1| + |l_2| + 2p_1 p_2!)}{w_1^2 w_2^2}} \frac{1}{2^{p_1 + p_2} |l_1 + l_2|!} \frac{\alpha_{|l_1 + l_2|} \gamma^{|l_1 + l_2|}}{2^{p_1 + p_2} |l_1 + l_2|! \gamma^{|l_1 + l_2|}} \exp \left[-\frac{\alpha^2}{4 \beta^2}\right]
\]

\[
\times \exp\left[-i \left(kr^2 + k\varphi + (l_1 + l_2)\varphi\right)\right] \sum_{j=0}^{\infty} \frac{c_j}{\beta^{2j}} \left(\frac{l_1 + l_2}{2} + j\right)! \left|\frac{l_1 + l_2}{2} + j\right|! \left(\frac{\alpha^2}{4 \beta^2}\right)^j
\]

where \(\alpha = kr / z\), \(\beta = \sqrt{(|l_1| + |l_2| + 1)!} + ik / 2z\), \(c_j\) denotes the coefficient of the series \(L_{p_1}^{l_1} (*) \times L_{p_2}^{l_2} (*)\). Now, if we assume \(w_1 = w_2 = w_0\), by substituting \(m = l_1 + l_2\) and \(n = (|l_1| + |l_2| - |l_1| + |l_2|)/2\) into Eq. (S3), we can reformulate the wave equation as

\[
E^{2\omega}(r, \varphi, z) = \frac{1}{\lambda z} \sqrt{\frac{2(|l_1| + |l_2| + 2p_1 p_2!)}{w_0^2}} \frac{1}{2^{p_1 + p_2} |l_1 + l_2|!} \frac{\alpha_{|l_1 + l_2|} \gamma^{|l_1 + l_2|}}{2^{p_1 + p_2} |l_1 + l_2|! \gamma^{|l_1 + l_2|}} \exp \left[-\frac{\alpha^2}{4 \beta^2}\right] \exp\left[-i \left(\frac{kr^2}{2z} + k\varphi + m\varphi\right)\right] L^{2\omega}(r, z)
\]

\[
E^{2\omega}(r, \varphi, z) = \sum_{j=0}^{p_1 + p_2} c_j \left(\frac{1}{\beta^{2j}} \left(n + j\right)! L_{n+j}^{l_1+l_2} \left(\frac{\alpha^2}{4 \beta^2}\right)\right) \frac{\partial_{z'}^{n+j}}{\partial_{z'}^{n+j}} \sum_{j=0}^{p_1 + p_2} q_j(z) L_{n+j}^{l_1+l_2} \left(\zeta_z\right)
\]

In the main text Eq. (S4) is abbreviated as \(E^{2\omega}(r, \varphi, z) = U(r, z; 2k, w_0, p_{1,2}, l_{1,2}) E^{2\omega}(r, z) \exp(-im\varphi)\) corresponding to Eq. (2).

At the far field, i.e., \(z \to z_R\), Eq. (S4) will become the Fourier transform of the SHG source and \(\zeta_z \to R\), which is given by
\[ \mathcal{F}[E^{2m}(\eta, \phi_0)] = \int_0^{2\pi} \int_0^\infty E_{\text{SHG}}(\eta, \phi_0) \exp[-i2\pi \eta r \cos(\phi - \phi_0)] r_0 dr_0 d\phi_0 \]
\[ = \frac{2^{2|m|} \pi_{p_1}^1 \pi_{p_2}^1}{(\pi_0^2 + p_1)(\pi_0^2 + p_2)!} \frac{1}{\alpha_0^{|m|}} \frac{1}{2\beta_0^{|m|}} \frac{1}{|z_0^2|^{\alpha_0^{|m|}}} \frac{1}{|z_0^2|^{\beta_0^{|m|}}} \frac{1}{4\beta_0^{|m|}} \exp\left( - \frac{\alpha_0^{|m|}}{4\beta_0^{|m|}} \right) \exp(-i\phi_0) L_{2m}(r, z_0) \]

\[ L_{2m}(r, z_0) = \sum_{j=0}^{\pi_{p_1}^1 \pi_{p_2}^1} q_j L_{2m,j}^m(\zeta) = \sum_{j=0}^{\pi_{p_1}^1 \pi_{p_2}^1} \left( c_{j} \frac{1}{\beta_0^{|m|}} (n + j)! \bar{L}_{2m,j}^m(\zeta) \right), \quad \text{for} \quad \ell_1 \times \ell_2 \geq 0 \]

where \( \zeta \) denotes a real function \( (\zeta \rightarrow \mathbb{R}) \). If we assume \( p_1 = p_2 \), the radial governing term, i.e., \( L_{2m}(r, z_0) \) can be further factorized as

\[ L_{2m}(r, z_0) = \begin{cases} L_{2m,j}^m(\zeta) \bar{L}_{2m,j}^m(\zeta), & \text{for} \quad \ell_1 \times \ell_2 \geq 0 \\ (p + n)! L_{2m,j}^m(\zeta) \bar{L}_{2m,j}^m(\zeta), & \text{for} \quad \ell_1 \times \ell_2 < 0 \end{cases} \]

**Appendix B  Additional Experimental Results**

**FIG. S1.** The comparison between the observed radial structure of the SHG of \( LG^2_p * LG^2_p \) with \( p = 0, 1, \) and \( 2 \), and the associated theoretical references.