Editors' Suggestion

Chiral relations and radial-angular coupling in nonlinear interactions of optical vortices

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In this article we investigate the connection between the chirality of interacting vortices and the appearance of radial structures in nonlinear wave mixing. Depending on the signs of their topological charges, the nonlinear mixing of optical vortices may produce a radial-angular coupling that generates a finite superposition of pure Laguerre-Gaussian modes carrying the resultant topological charge and a finite spectrum of radial orders. These radial modes evolve with different Gouy phases that determine the transformation from a hollow intensity distribution in the near field to a finite ring structure in the far field pattern. In this sense, we interpret the appearance of radial modes in nonlinear wave mixing as a diffraction of the up-converted beam through the effective amplitude-phase mask created by the pump beams. This interpretation is supported by comparison between the images produced by the nonlinear process and purely diffractive measurements with a spatial light modulator that mimics the amplitude-phase modulation produced in nonlinear wave mixing.

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I. INTRODUCTION

The cross-talk between different optical degrees of freedom is a central issue for classical and quantum communication protocols where controlled operations are required [1–5]. Polarization controlled mixing of orbital angular momentum (OAM) in nonlinear processes has been investigated in recent works [6–9]. Much progress in this field was made possible due to improved techniques employing spatial light modulators (SLMs) [10]. The spatial intensity distribution and the quantum correlations under polarization control in nonlinear media have been investigated [6,11,12]. The cross-talk between radial and angular modes has been considered in connection with entanglement and teleportation schemes [13].

Parametric processes driven by vortex beams can generate radial structures depending on the relative chirality of the interacting vortices. The appearance of radial structures in nonlinear processes has been observed in different contexts [14–17]. In second harmonic generation (SHG) the nonlinear mixing of Laguerre-Gaussian (LG) modes occurs under nontrivial selection rules involving radial and angular indices [7,18,19]. In this case, when counter-rotating vortices are nonlinearly mixed, the resulting topological charge in the second harmonic does not match the accompanying radial power law. This radial-angular mismatch results in a transverse structure that does not preserve its shape along free propagation, evolving from a hollow intensity distribution in the near field to a ring structure in the far field. In this work we investigate the diffractive origin of radial structures generated in nonlinear vortex interactions. First, we show that a radial-angular mismatched beam can be cast as a finite superposition of radial modes and the corresponding radial spectrum is analytically derived. Then, we use a spatial light modulator (SLM) to mimic the amplitude-phase modulation produced in SHG and compare it with the actual structure generated by the nonlinear process. The experimental results confirm our theoretical derivation of the radial mode spectrum and support the diffractive interpretation. This can be relevant in different physical contexts such as nonlinear wave mixing in atomic media [20], high harmonic generation (HHG) [21–23], and self-phase modulation in polariton superfluids [24,25], for example.

II. THE RADIAL SPECTRUM OF NONLINEAR WAVE MIXING WITH LAGUERRE-GAUSSIAN MODES

Two-dimensional Laguerre-Gaussian (LG) functions are well known solutions of the paraxial wave equation in cylindrical coordinates, where the longitudinal coordinate z designates the propagation direction and (r, ϕ) are the polar coordinates in the transverse plane. In a compact form, the normalized LG modes can be expressed as

$$\psi_{pl}(\tilde{r}, \phi, \tilde{z}) = \mathcal{N}_{pl}\, (\sqrt{2}\,\tilde{r})^{|l|}\, L_p^{|l|}(2\tilde{r}^2)\, e^{-(1+i\tilde{z})\tilde{r}^2} \times e^{\left[il\phi + i(2p+|l|+1)\arctan(\tilde{z})\right]}, \tag{1}$$

where $L_p^{|l|}(x)$ is a generalized Laguerre polynomial and

$$\mathcal{N}_{pl} = \sqrt{\frac{2\,p!}{\pi\, w^2(\tilde{z})(p+|l|)!}},$$
$$w(\tilde{z}) = w_0\sqrt{1+\tilde{z}^2},$$
$$\tilde{r} = r/w(\tilde{z}),\ \tilde{z} = z/z_0. \tag{2}$$

$z_0 = k\, w_0^2/2$ is the Rayleigh distance and w_0 is the beam waist. The LG modes form an orthonormal and complete basis of square-integrable functions in the transverse coordinates, which can be expressed as

$$\int \psi_{pl}^*(\tilde{\mathbf{r}},\tilde{z})\,\psi_{p'l'}(\tilde{\mathbf{r}},\tilde{z})\,d^2\tilde{\mathbf{r}} = \delta_{pp'}\,\delta_{ll'},$$
$$\sum_{p,l}\psi_{pl}^*(\tilde{\mathbf{r}},\tilde{z})\,\psi_{pl}(\tilde{\mathbf{r}}',\tilde{z}) = \delta(\tilde{\mathbf{r}}-\tilde{\mathbf{r}}'). \tag{3}$$

These modes are stable under free propagation as they preserve their functional dependence on the transverse coordinates, except for global phase factors and an expanding beam width $w(\tilde{z})$. This stability requires a radial-azimuthal match given by the parameter l that appears on both phase and amplitude terms. When this match is violated, the whole structure loses stability and the resulting function changes its shape along propagation [26]. As we will show, it can be decomposed as a superposition of radial orders that evolve with different Gouy phases, which explains the near-to-far field transformation [27]. This fact is of great significance for nonlinear mixing of optical vortices as it naturally generates radial-azimuthal mismatch. We next derive the radial spectrum of such mismatched structures in terms of pure LG modes.

The propagation properties of hollow beams without OAM have been discussed in [28,29]. This is a special case of radial-angular mismatch that appears, for example, when opposite topological charges ($\pm l$) are added in three-wave mixing, resulting in null OAM. Here we study the more general situation involving nonlinear mixing of independent topological charges l_1 and l_2 in type-II second harmonic generation (SHG) [7,8]. We consider the $p=0$ fundamental beam, which yields a Laguerre polynomial equal to 1 and the Gouy phase is zero at $z=0$. The resulting field distribution at the output of the nonlinear crystal ($z=0$) is of the form

$$\begin{aligned}\left(\psi_{0l_1}\,\psi_{0l_2}\right)_{z=0} &= \mathcal{N}_{0l_1}\,\mathcal{N}_{0l_2}\,(\sqrt{2}\,\tilde{r})^{|l_1|+|l_2|}\,e^{-2\tilde{r}^2}\,e^{i(l_1+l_2)\phi}\\ &= \frac{\mathcal{N}_{0l_1}\,\mathcal{N}_{0l_2}}{2^{l/2}\,\mathcal{N}_{0l}}\,\zeta_{0lm}(\sqrt{2}\,\tilde{r},\phi,0),\end{aligned} \tag{4}$$

where we defined the normalized structure

$$\zeta_{0lm}(\tilde{r},\phi,0) = \mathcal{N}_{0l}\,(\sqrt{2}\tilde{r})^l\,e^{-\tilde{r}^2}\,e^{im\phi}, \tag{5}$$

with $l=|l_1|+|l_2|$ and $m=l_1+l_2$. When corotating vortices are mixed ($l_1l_2 \geqslant 0$), the radial and azimuthal indexes match ($|m|=l$) and ζ_{0lm} is a pure LG mode ψ_{0m} with zero radial order ($p=0$) and rescaled width. However, when counter-rotating vortices are mixed ($l_1l_2<0$), the net OAM left in the up-converted beam does not match the radial power law ($|m|\neq l$). In this case (5) is no longer a solution of the paraxial equation, and the beam will not be self-similar upon propagation, but can still be decomposed on its natural LG modes in the form

$$\zeta_{0lm}(\tilde{r},\phi,0) = \sum_p \mathcal{C}_{pm}\,\psi_{pm}(\tilde{r},\phi,0), \tag{6}$$

involving different radial orders and a fixed topological charge m. Equation (6) describes the near field ($z=0$) distribution, so the far field behavior is readily obtained by including the z-dependent phase factors associated with the quadratic and Gouy phases.

In order to derive the coefficients $\mathcal{C}_{pm}$, it will be useful to write Eq. (6) in the form

$$\zeta_{0lm}(\tilde{r},\phi,0) = \mathcal{N}_{0l}\,(\sqrt{2}\tilde{r})^{|m|}\,e^{-\tilde{r}^2}\,e^{im\phi}\,(2\tilde{r}^2)^P, \tag{7}$$

where $P \equiv (l-|m|)/2$. Here, a subtle feature shows up. When $P\in\mathbb{N}$, the monomial can be expanded as a finite sum of generalized Laguerre polynomials $L_p^k(x)$,

$$x^P = \sum_{p=0}^{P}\frac{(-1)^p\,P!\,(k+P)!}{(P-p)!\,(k+p)!}\,L_p^k(x) \quad (P\in\mathbb{N}). \tag{8}$$

Note that the summation runs over the index p, but the family k is arbitrary and can be chosen at will, so long as the adequate coefficients are used. By making $x \equiv 2\tilde{r}^2$ and choosing $k=|m|$, Eq. (7) can be cast as a finite superposition of LG modes with different radial orders $0\leqslant p\leqslant P$ and a fixed topological charge m. Including the quadratic and Gouy phase factors, the LG expansion valid in both the near and far fields becomes

$$\zeta_{0lm}(\mathbf{r}) = \frac{P!Q!}{\sqrt{(P+Q)!}}\sum_{p=0}^{P}\frac{(-1)^p\,\psi_{pm}(\mathbf{r})}{(P-p)!\sqrt{p!(Q-P+p)!}}, \tag{9}$$

where $Q=(l+|m|)/2$. The radial-angular mismatch P determines the dimensionality of the radial spectrum. When $P\notin\mathbb{N}$, i.e., when it is either negative or half-integer, the radial spectrum is infinite. Interestingly, nonlinear OAM mixing always generates finite radial spectra, since it involves products of LG modes, implying

$$\begin{aligned} P &= \frac{|l_1|+|l_2|-|l_1+l_2|}{2} = \min\left(|l_1|,|l_2|\right)\in\mathbb{N},\\ Q &= \frac{|l_1|+|l_2|+|l_1+l_2|}{2} = \max\left(|l_1|,|l_2|\right)\in\mathbb{N}.\end{aligned} \tag{10}$$

Therefore, the product of counter-rotating LG modes results in a finite radial spectrum with $\min(|l_1|,|l_2|)+1$ terms and a rescaled width. Without loss of generality, we shall assume $|l_1|\leqslant|l_2|$. Then, the LG decomposition of the mode product reads

$$\psi_{0l_1}(\tilde{\mathbf{r}})\psi_{0l_2}(\tilde{\mathbf{r}}) = \sqrt{\frac{2}{\pi w^2}}\sum_{p=0}^{|l_1|} C_{p,l_1+l_2}\,\psi_{p,l_1+l_2}(\sqrt{2}\tilde{\mathbf{r}}), \tag{11}$$

where

$$C_{p,l_1+l_2} = \frac{(-1)^p}{(|l_1|-p)!}\sqrt{\frac{|l_1|!|l_2|!}{2^{|l_1|+|l_2|}p!(|l_2|-|l_1|+p)!}}. \tag{12}$$

Equations (11) and (12) can be easily generalized for products involving modes with different waists, as was done in Ref. [7]. We next compare the experimental results obtained with nonlinear mixing of counter-rotating vortices with those given by the spatial modulation from a SLM programed with a LG mode product.

III. EXPERIMENTAL RESULTS

The experimental setup is sketched in Fig. 1. First, we obtained images from nonlinear mixing of two optical vortices

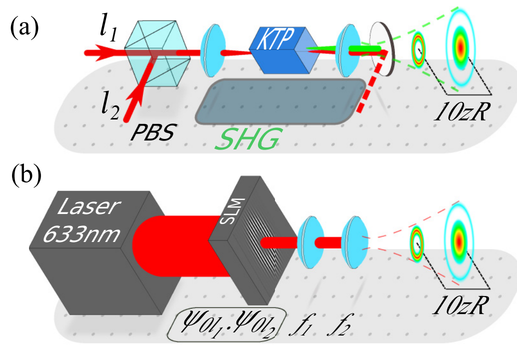

FIG. 1. (a) Experimental scheme for nonlinear mixing of different topological charges in second harmonic generation. (b) Preparation of radial-angular mismatch from Laguerre-Gaussian mode product with a spatial light modulator (SLM).

with topological charges l_1 and l_2 in second harmonic generation (SHG) as shown in Fig. 1(a). Two vortices with orthogonal polarizations at 1064 nm wavelength are combined in a polarizing beam splitter (PBS) and sent to a potassium titanyl phosphate (KTP) crystal where SHG takes place under type-II phase match. A new vortex beam is generated at 532 nm and separated from the fundamental frequency by a dichroic glass. The near-to-far field transition is studied by imaging the second harmonic beam with a charge-coupled device camera (CCD Imaging Source model DMK31BF03.H). Then, the SHG images are compared with those generated by diffraction of an input plane wave on a spatial light modulator (SLM), as shown in Fig. 1(b). A 633 nm continuous wavelength He-Ne laser is expanded to a 4 mm wide Gaussian beam that impinges on the SLM (Hamamatsu LCOS-SLM model XT10468) programed with the amplitude-phase modulation given by the product of Laguerre-Gaussian functions [10]. We want to demonstrate the generation of radial modes solely from the OAM combination, so we will consider only products involving zero radial order. The transmission function programed on the SLM plane ($z = 0$) will be of the form

$$T(\tilde{r}, \phi) = T_0\, \tilde{r}^{(|l_1|+|l_2|)}\, e^{-2\tilde{r}^2}\, e^{i(l_1+l_2)\phi} \propto \psi_{0l_1}\, \psi_{0l_2}, \qquad (13)$$

where T_0 is a positive real factor. Although a reflective SLM was employed, we draw a transmission scheme in Fig. 1(b) for easier comparison with the SHG measurements. The beam generated by the SLM is sent through two imaging lenses with focal lengths $f_1 = 10$ cm and $f_2 = 15$ cm, placed at 70 and 100 cm from the SLM, respectively. This produces a short Rayleigh range ($z_R \approx 0.66$ mm) allowing capture of the near-to-far field transition over a short propagation distance. The CCD camera is displaced between 150 and 155 cm from the SLM, where the near and far field patterns are registered. Note that the amplitude modulation depends only on $|l_1| + |l_2|$ and is completely insensitive to the relative chirality in the mode product, while the phase modulation depends on $l_1 + l_2$, giving different results for co- and counter-rotating charges. Since we want to put into evidence the role of the relative chirality in the generation of higher radial orders, we will consider only products with $l_1\, l_2 < 0$ and compare with the corresponding experimental results given by the actual nonlinear mixing of the same topological charges. In all cases,

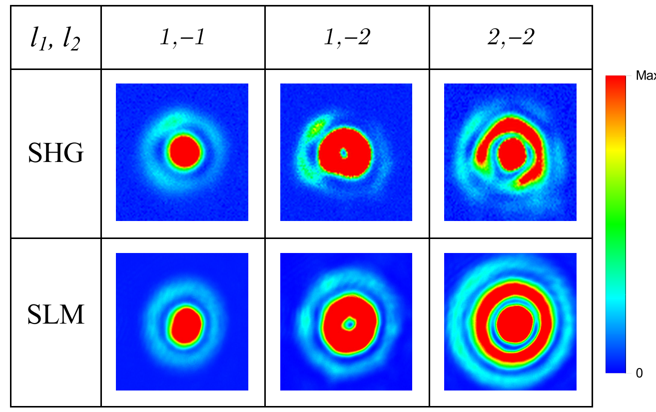

FIG. 2. Comparison between the far field images obtained with OAM addition in SHG (top) and those with the SLM (bottom) programed with products of LG modes. Different combinations of counter-rotating topological charges were employed, showing excellent agreement in all cases.

the near-to-far field transition reveals the radial orders through external rings that become observable. We consider different combinations of counter-rotating vortices, as shown in Fig. 2. According to the radial spectrum given by Eq. (9), we expect that min $(|l_1|, |l_2|)$ radial orders appear in the transmitted field. In all cases, the radial orders generated can be easily identified by the number of outer bright rings in the far field. Moreover, one can notice the presence of the phase singularity in all the cases where there is a net topological charge ($l_1 + l_2 \neq 0$). The agreement between the images produced by nonlinear mixing and those obtained by the SLM modulation confirms the diffractive origin of the radial structure. In nonlinear processes driven by LG modes, the pump beams produce a spatial modulation of the up-converted frequency with LG mode products that may result in radial-azimuthal mismatch, giving rise to a radial spectrum.

The role of the radial-angular mismatch can be further evidenced by testing the far field images obtained from the SLM modulation with different (l_1, l_2) combinations having the same radial power law variation $|l_1| + |l_2| = 6$, as shown in Fig. 3. The theoretical images are obtained from the Fresnel

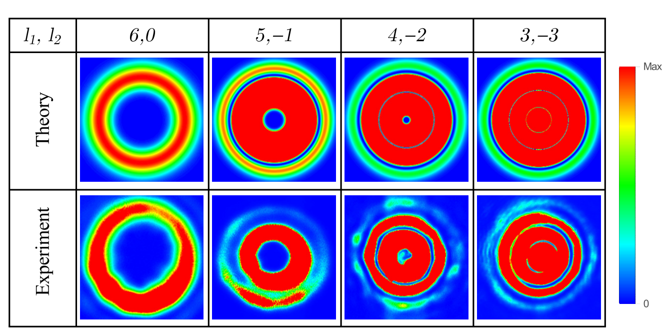

FIG. 3. Comparison between numerical simulations (top) and experimental results (bottom) of the diffraction pattern obtained with $|l_1| + |l_2| = 6$ and $0 \leqslant l_1 + l_2 \leqslant 6$. Saturation in both numerical and experimental images was intentional in order to make clear the external rings.

diffraction integrals, with excellent agreement with the experimental results given by the SLM modulation.

We can extend our argument to *arbitrary* nonlinear processes. The response of a nonlinear medium to an incident electromagnetic field is of a general form

$$\mathbf{P}^{(n+m)} = \chi^{(n+m)}\,\mathbf{E}_1\cdots\mathbf{E}_n\,\mathbf{E}^*_{n+1}\cdots\mathbf{E}^*_{n+m}, \tag{14}$$

where $\chi^{(n+m)}$ is the nonlinear susceptibility tensor and $\mathbf{P}^{(n+m)}$ is the nonlinear polarization of the medium corresponding to n up-conversion and m down-conversion steps. We want to describe the nonlinear generation of radial orders, so we assume the input field to be a superposition of different OAM components, all carrying zero radial orders. For simplicity, we assume a collinear configuration along z, with a thin nonlinear medium placed at the focal region ($z = 0$) of the incoming beams. Then, the input field can be written as

$$\mathbf{E}_{in} = \sum_l \mathbf{A}_{0l}\,\psi_{0l}(\tilde{r},\phi,0)\,e^{-i\omega t}, \tag{15}$$

giving rise to nonlinear polarization contributions proportional to a product of LG modes of the form

$$P^{(n+m)}_{l_1\cdots l_{n+m}} \propto \psi_{0l_1}\cdots\psi_{0l_n}\psi^*_{0l_{n+1}}\cdots\psi^*_{0l_{n+m}}. \tag{16}$$

So long as the nonlinear interaction is restricted to the focal region of the beams, complex conjugation amounts only to a topological charge inversion ($\psi^*_{0l} \rightarrow \psi_{0,-l}$) in the LG mode product. The transverse modes generated in the nonlinear process will depend on the LG decomposition of such product contributions. They may result in radial-angular mismatched field distributions of the form ζ_{0LM}, with

$$\begin{aligned} L &= \sum_{j=1}^{n+m}|l_j|,\\ M &= \sum_{j=1}^{n} l_j - \sum_{j=n+1}^{n+m} l_j. \end{aligned} \tag{17}$$

The appearance of radial orders in the nonlinear process will be subjected to the mismatch between the radial power law L and the net topological charge M. Strong indications of this mechanism can be found in the images produced by four-wave mixing in atomic vapors shown in [20]. This can also be relevant for self-phase modulation in both polariton superfluids [24,25] and saturable optical media [30]. For example, a generalized product can be written as

$$\prod_{j=1}^{n}\psi_{0l_j}\prod_{j=n+1}^{n+m}\psi^*_{0l_j} = \frac{\prod_{j=1}^{n+m}\mathcal{N}_{0l_j}}{(n+m)^{l/2}\,\mathcal{N}_{0L}}\,\zeta_{0LM}(\sqrt{n+m}\,\tilde{\mathbf{r}}), \tag{18}$$

with L and M given by Eqs. (17). We can deduce the dimension of the radial spectrum by identifying the positive and negative contributions to the net topological charge M. Let $l_\pm$ be the sum of positive (negative) contributions to the net topological charge. Then, we have $L = |l_+| + |l_-|$, $M = l_+ + l_-$. The dimension of the radial spectrum generated by the multimode product is simply $P = \min(|l_+|, |l_-|)$.

IV. CONCLUSION

In conclusion, we demonstrate the connection between the relative chirality of the interacting beams and the appearance of radial structures in nonlinear wave mixing. The stability of a vortex structure along propagation depends on the match between the vortex topological charge and the radial power law variation of the field amplitude. When this condition is violated, a radial spectrum is created and a ring structure is formed in the far field. This radial-azimuthal mismatch is naturally produced in nonlinear wave mixing, where the appearance of radial orders is subjected to the relative chiralities of the interacting beams. We have confirmed this effect with both three-wave mixing and pure spatial modulation of an expanded Gaussian beam by a SLM programed with products of Laguerre-Gaussian functions. The radial orders generated agree with our theoretical derivation. This effect can be important to other nonlinear phenomena and possibly in the quantum domain, where the number of excited modes determines the dimension of the photonic Hilbert space. This opens interesting perspectives for future investigations on the subject.

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