Directly Determining Orbital Angular Momentum of Ultrashort Laguerre–Gauss Pulses via Spatially-Resolved Autocorrelation Measurement

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Autocorrelation measurement based on second-harmonic generation (SHG), the best-known technique for measuring the temporal duration of ultrashort pulses, dates back to the birth of ultrafast lasers. Here, it is proposed and experimentally demonstrated that such well-established technique can be used to measure the orbital angular momentum of ultrashort Laguerre–Gauss (LG) pulses. By analyzing the spatial spectrum (i.e., the far-field pattern) of the SHG signal with a spatially-resolved detector, the full spatial structure of ultrashort LG pulses, including both azimuthal and radial indices, are unambiguously determined. The results provide an important advancement for the well-established autocorrelation technique by extending it to reach its full potential in laser characterization, especially for structured ultrashort pulses.

1. Introduction

Ultrashort pulses carrying orbital angular momentum (OAM) have gained an increasing interest in the community of ultrafast photonics and structured light.[1–9] An important reason for this is that optical vortices with ultrahigh peak power and ultrashort duration provide a new tool to explore novel applications, as well as fundamental aspects of physics. For instance, ultrashort vortex pulses structured in their spatial or polarization degrees of freedom, can produce light fields with extraordinary spatiotemporal and topological structures.[10–14] In a similar way, high-order harmonic generation of ultrashort vortex pulses can provide a wide variety of novel light-matter interactions.[15–19] Importantly, most of the ultrashort vortex pulses in the aforementioned studies focus only on Hypergeometric-Gaussian vortex modes embedded with OAM, which are not eigenmodes of the paraxial-wave equations (PWE) with propagation-invariant transverse structure in free space.[20] For a further study it is required to consider OAM modes with well-defined radial structure, such as Laguerre–Gaussian (LG) modes, which are indeed solutions of the PWE in cylindrical coordinates.[21] Importantly, such propagation-invariant modes play an important role in various applications.[22,23] It is therefore not surprising that the generation and applications of ultrashort pulsed LG modes have aroused high interest in recent time.[2,24] As such, a proper characterization of these modes is curial in various research fields.

In this way, a full characterization of LG modes requires the simultaneous determination of their azimuthal and radial indices. A common way to accomplish this task involves mode-projective measurement, which requires the projection of the unknown LG mode onto several elements of the LG basis, which is usually implemented through spatial light modulators.[25,26] Importantly, this commonly used technique requires a precise priori knowledge of some of the parameters of the unknown beam, such as its initial beam width radius $w_0$ and its radius of curvature $R_z$, for an accurate characterization of the mode in question.[27–29] For ultrashort pulses, transverse dispersion induced by the grating of complex-amplitude modulation would bring additional issues. Although a recent work has suggested that one can measure OAM of intense ultrashort pulses by exploiting strong-field ionization,[30] a simple technique towards commercial uses has not been developed.

More recently, the idea of the mode-projective measurement was used with nonlinear interactions and applied to numerous novel concepts, such as nonlinear modal detection and spatial mode teleportation,[31,32] enriching the already active topic of structured nonlinear optics.[30–40] The history of ultrashort-pulse characterization with nonlinear optics dates back to the birth of ultrafast lasers. Perhaps one of the most known techniques is the so-called autocorrelation technique based on second-harmonic generation (SHG) that is widely used to measure the temporal duration of ultrashort pulses.[41] In principle, the autocorrelation method is easy to implement but due to its time-integrating detection losses the complex-amplitude information of the electric
field. Importantly, to observe the longitudinal structure of an optical pulse, previous works have used the spectrum-resolved detection to further improve the autocorrelation technique, which has been termed, frequency-resolved optical gating, commonly known as FROG. In this manuscript, inspired by this revolutionary idea, we bring into play the transverse properties of light. More precisely, by introducing a spatially-resolved detection, we extend the well-established autocorrelation technique to the characterization of the spatial structure of ultrashort pulses, where the simplicity of the technique is the main feature. Hence, by theoretical analysis and experimental verification, we show that the spatial spectrum (i.e., the far-field pattern) of the SHG is equivalent to the autocorrelation function of spatial modes and thus, in addition to measuring its pulse width, it can also be used to determine the OAM content of ultrashort pulsed LG modes.

2. Experimental Section

Laguerre–Gaussian modes represent a well-known family of paraxial beams featuring a ring-shaped (or donut-like) intensity pattern, except for those with a topological charge \( \ell = 0 \). As eigen solutions of the PWE, any LG mode, or modes resulting from superpositions of the same modal order \( N \), maintained their transverse pattern upon propagation (termed self-similar beams), apart from a diameter scaling, \( w_z = w_0 |1 + (2z / k w_0)^2|^{1/2} \), where \( w_0 \) is the beam radius at the waist plane. The spatial complex amplitude of an LG mode in cylindrical coordinates \( \{r, \varphi, z\} \) has the form,

\[
\begin{align*}
LG_p^\ell(r, \varphi, z) &= \sqrt{\frac{2p!}{\pi (p + |\ell|)!}} \frac{1}{w_z} \left( \frac{\sqrt{2r}}{w_z} \right)^{|\ell|} L_p^{|\ell|} \left( \frac{2r^2}{w_z^2} \right) \exp \left( \frac{-r^2}{w_z^2} \right) \\
&\quad \times \exp \left\{ -i \left[ \frac{kz}{2R_z} + \ell \varphi - (N + 1) \tan^{-1} \left( \frac{z}{z_R} \right) \right] \right\},
\end{align*}
\]

where \( \ell = 0, \pm 1, \pm 2, \ldots \) and \( p = 0, 1, 2, \ldots \) are the azimuthal (topological charge) and radial indexes, respectively. The function \( L_p^{|\ell|} \) is the associated Laguerre polynomial, which determines the radial amplitude structure of the mode in the transverse plane, and \( N = 2p + |\ell| \) defines its modal order. The parameters \( z_R = k w_0^2 / 2 \) and \( R_z = z^2 + z_R^2 / z \) are the Rayleigh length and radius of curvature, respectively. The factor \( \exp(\ell \varphi) \) in Equation (1) is an eigenfunction of the operator \(-i \partial_\varphi = -i(\partial_r + r \partial_r)\); that is, LG modes with \( \ell \neq 0 \) are natural carriers of optical OAM and the associated eigenvalue \( \ell \) is the OAM quantum number. Moreover, because optical OAM originates from twisted wavefronts involving phase singularities, the azimuthal index is also known as “topological charge”.

To fully characterize an unknown LG mode, its azimuthal (\( \ell \)) and radial (\( p \)) indices must be determined simultaneously. It is noteworthy that the latter can be determined directly by counting the number of rings or dark nodes in the ring-shaped pattern of an LG mode, as shown in the examples in Figure 1c. Therefore, the main task was to determine the azimuthal index (or topological charge \( \ell \)) of an unknown LG mode through an autocorrelation measurement based on SHG, which is usually used to characterize the temporal duration of ultrashort pulses. To study the autocorrelation in the spatial degree of freedom, the SHG of two conjugated LG modes was considered, whose spatial complex amplitude at the generation plane (usually inside the crystal) is given by

\[
E^{2n}(r, \varphi, z_0) = LG_p^\ell(r, \varphi, z_0) LG_p^{-\ell}(r, \varphi, z_0)
\]

\[
= u^{2n}(r, z_0) L_p^{|\ell|} \left( \frac{2r^2}{w_z^2} \right) L_p^{|\ell|} \left( \frac{2r^2}{w_z^2} \right),
\]

where \( u^{2n}(r, z_0) \) is the amplitude envelope of SHG at the generation plane. Hence, the spatial spectrum of the SHG can be derived through the Fourier transformation of Equation (2), which is equivalent to the spatial complex amplitude at the far field, namely,

\[
E^{2n}(r, \varphi, z_0) = F \left[ E^{2n}(r, \varphi, z_0) \right]
\]

\[
= u^{2n}(r, z_0) \rho(\zeta),
\]

where \( u^{2n}(r, z_0) \) is the amplitude envelope at the Fourier plane (far field), \( \zeta \) is a real function associated with scaling factors of the Fourier lens, and \( \rho(\zeta) \) governs the far-field radial structure that can be further factorized as (see Ref.[37] for more details)

\[
\rho(\zeta) = \frac{(p + |\ell|)!}{p!} L_p^{|\ell|}(\zeta) L_p^{|\ell|}(\zeta), \quad \text{for} \quad \ell \neq 0,
\]

\[
\rho(\zeta) = L_p^{|\ell|}(\zeta) L_p^{|\ell|}(\zeta), \quad \text{for} \quad \ell = 0.
\]

The general case \( \ell \neq 0 \), which is given in Equation (4), was first considered. Crucially, here the product \( L_p^{|\ell|}(\zeta) L_p^{|\ell|}(\zeta) \) contained \( n = 2p + |\ell| = N \) zeros, as shown in Figure 1a, which means there were \( N \) phase jumps in the far-field wavefront along the radial direction. Due to the zero amplitude at the phase jumps, \( N \) dark nodes occurred in the far-field pattern of the SHG. Hence, the azimuthal index of an LG mode can be directly determined from its spatial spectrum (i.e., far-field autocorrelation pattern), namely, by using the relation \( |\ell| = N - 2p \), where the index \( p \) is known directly from the dark-node number of fundamental-frequency pattern. For the second special case, it was seen that the term \( L_p^{|\ell|}(\zeta) L_p^{|\ell|}(\zeta) \) in Equation (5) is the same as that in Equation (2) when \( \ell = 0 \). Thus, the radial structure of the SHG beam in the far field was the same as that in the generation plane, in which \( \rho(\zeta) \) had \( n = p \) zeros, as shown in the example of Figure 1b. In other words, the pattern of SHG in this special case was just the square of the input LG-mode pattern and undergoes self-imaging from the near field \( \{z_0\} \) to the far field \( \{z_\infty\} \) (see Supporting Information for details).

Overall, the number of dark nodes \( n \) in the spatial spectrum of or far-field autocorrelation patterns, which can be used to determine the LG mode in both cases, is given as

\[
\begin{align*}
n &= 2p + |\ell| \quad \text{for} \quad \ell \neq 0 \\
&\quad n = p \quad \text{for} \quad \ell = 0.
\end{align*}
\]

To exemplify this, Figure 1c shows the spatial complex amplitude of LG modes for different azimuthal and radial indices, and Figure 1d shows their corresponding spatial autocorrelation, that...
Figure 1. Theoretical results: a,b) simulated $\rho(\zeta)$ curves for autocorrelations of $\text{LG}_{12}$ and $\text{LG}_{02}$, respectively. c,d) Simulated beam profiles of LG modes and their corresponding spatial autocorrelations, respectively, where patterns in (d) are drawn with saturated intensity. The phase profiles of (c) and (d), and detailed radial structure of (d) are given in Supporting Information.

is, their SHG patterns at the far field. Here, to clearly show the dark nodes, the beam profiles in Figure 1d were drawn with saturated intensity. Notice that, i) for the common case $\ell \neq 0$, one can directly measure the radial index $p$ and modal order $N$ of LG modes from the dark-node numbers in their beam intensity patterns ($p$) and associated autocorrelation patterns ($n$), as shown in Figures 1c, and 1d, respectively, and thus can further determine their topological charges via the relation $|\ell| = n - 2p$. ii) For the special case $\ell = 0$, the radial index $p$ was recognized from the dark-node numbers in their autocorrelations patterns, which actually were the same as the dark-node numbers in the original beam profiles shown in Figure 1 (i.e., $n = p$).

In the following, the above theory was verified experimentally. Figure 2 shows a schematic diagram of the experimental setup, in which a collinear autocorrelation framework based on type-II SHG was used. More specifically, the signal source was an ultrafast laser operating at a wavelength of 800 nm and with a repetition rate of 80 MHz. The input laser beam was first prepared into the desired LG mode by using a complex-amplitude hologram displayed on a spatial light modulator. The prepared LG mode was then set to a diagonally polarized state and sent afterwards to a polarizing Michelson interferometer. In one arm, a mirror mounted on a translation stage was used to control the pulse delay and, in the other arm, a cylindrical lens was used to conjugate the OAM of the input LG mode. As a result, the output beam from the interferometer was a partial cylindrical vector beam, that is, $1/\sqrt{2}[\text{LG}_{\ell p}(t)\hat{e}_H + \text{LG}_{-\ell p}(t - \tau)\hat{e}_V]$. Here the term “partial” means that the cylindrical vector mode was not necessarily in a pure vector state due to the delay $\tau$. The output cylindrical vector mode was then focused into a 0.5 mm $\beta$-$\text{BaB}_2\text{O}_4$ (BBO) crystal with type-II phase matching to generate the SHG (i.e., autocorrelation signal). Finally, the autocorrelation signal was focused by a Fourier lens onto a CMOS-based camera, where the spatial spectrum of the SHG was recorded.

The inset at the right-bottom of Figure 2 shows the measured temporal autocorrelation signal, that is, $I^{2w}(t) = \int I(\tau)I(\tau - t)dt$, where a 54.5 $\mu$m full width at half maximum (FWHM) indicated that the temporal duration of the input pulses was $\approx 128$ fs. Regarding this collinear SHG configuration, it is important to note that, even if the polarizing extinction ratio of the type-II SHG was not good enough, a great signal to noise ratio can still be obtained. Because the temporal signal was obtained...
by adding the intensities of the pixels in the central region of the SHG pattern (see the top-right inset of the bigger bottom-left inset of Figure 2), that is, the Gaussian beam enclosed by dashed ring, or rather, the energy in the central region was completely attributed to the ‘sum-frequency generation’ of a mutually conjugate LG-mode pair. Afterward, the path delay was fixed at an appropriate position so that a maximum intensity of the temporal signal can be obtained, the spatial autocorrelation signal (i.e., the far-field SHG pattern) was then recorded with a camera. Figures 3a, and 3b show the measured LG modes with different modal indexes ($\ell$ and $p$) and their corresponding spatial autocorrelations, respectively. These results showed that, for the common case $\ell \neq 0$, the dark-node number in the far-field SHG pattern was exactly the same as the measured modal order of LG modes (i.e., $n = 2p + |\ell| = N$). For the special case, the observed autocorrelation patterns of LG modes with $\ell = 0$ were exactly the same as the square of their beam profiles shown in Figure 3a (i.e., $n = p$).

In addition, note that as shown in Figure 1d, the intensity ratio between the center and the surrounding regions of spatial-spectrum patterns usually exceeded the 8-bit dynamic range. Therefore, the spatially-resolved detection required a high dynamic range (HDR) to exactly measure the spatial spectrum of SHG. To solve this issue, there were two feasible choices: the straightforward way was using a scientific camera with 12-bit or higher dynamic range but high cost; the alternative way was to use a common commercial camera, for example, low-cost CMOS sensors, and combine this with integral-time multiplex to realize the HDR detection. The second one was preferred for most cases, and a low-cost CMOS with an 8-bit dynamic range was also used. Besides, all the autocorrelation patterns shown in Figure 3b were overexposed to see the dark-node numbers clearly. The detailed radial structure of the patterns is provided in the Supporting Information.

3. Discussion

We experimentally verified that the spatial-resolved autocorrelation measurement allows to characterize both the temporal and spatial properties of high-order LG beams. One reason for obtaining this important result is that the spatial spectrum derived from Equations (2) and (3) is mathematically equivalent to the optical autocorrelation functions of LG modes, which can be used to determine the topological charge of both the coherent and partial coherent high-order LG modes without requiring a priori knowledge.\(^{43,44}\) Alternatively, the results can be interpreted by considering the radial-mode selection rule of LG modes in SHG and the evolution of the Gouy-phase-mediated pattern of superposed LG modes.\(^{37,45,46}\) For the sake of clarity, we included additional results as Supporting Information, related to the
structural evolution of the SHG beams from the near to the far field. In addition, it is also worth mentioning that this interesting phenomenon in the case $\ell = 0$ (i.e., self-imaging of SHG from the near field to the far field) actually exists in a more general sum-frequency generation of two LG modes $LG(\ell_1, p_1) \ast LG(\ell_2, p_2)$, in which $\ell_1 \times \ell_2 \geq 0$ and $p_1 = p_2$ (see ref. [37] for further details).

Note that interferometric wavefront detection is also often used in measuring OAM of beams, but does not work well for ultrashort pulses. The reason is that the visibility of interference between reference (or probe) beam and ultrashort pulses, which depends on the duty cycle of the pulses, is extremely low. The technique demonstrated above can be seen as an advanced autocorrelator, which maintains the simplicity structure of its predecessor version. More importantly, this technique exploits the spatial degree of freedoms of the parametric field to characterize the spatial structure of ultrashort pulses. At this point, a question worth asking is, can we use the spatial-resolved detection to characterize the fine longitudinal structure of light fields? After all, the frequency-resolved detection used in FROG is usually more expensive and its performance is difficult to improve. A possible answer lies in the phase spectrum within the spatial modes (or intramodal phase), which provides potential opportunity to observe the electric-field amplitude at a subwavelength scale.\(^{[47]}\) Hence, exploiting the sum-frequency generation between the measured pulse and a reference beam with known spatial mode holds a great potential.

In summary, we have theoretically proposed and experimentally verified that the autocorrelation measurement, beyond characterizing the temporal structure of light, can also be used to determine the spatial structure of LG modes, including both the azimuthal and radial indices. More specifically, to determine an unknown LG beam, we first determine the radial index by counting the dark-node number $n$ in its original beam profile. Next, if the beam carries nonzero topological charge (i.e., whether a Gaussian mode exists in the center of the beam profile), we can further determine the modal number $N$ by counting the dark-node number $n$ in the far-field autocorrelation pattern (i.e., SHG spatial spectrum), which gives its azimuthal index via the relation $|\ell| = n - 2p$. These results expand the present nonlinear autocorrelation technique, allowing it to reach its full potential in laser characterization in this era of structured light.\(^{[48-50]}\)

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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