Effects of the Coupling between the Orbital Angular Momentum and the Temporal Degrees of Freedom in the Most Intense Ring of Ultrafast Vortices

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Abstract: It has recently been shown that the temporal and the orbital angular momentum (OAM) degrees of freedom in ultrafast (few-cycle) vortices are coupled. This coupling manifests itself with different effects in different parts of the vortex, as has been shown for the ring surrounding the vortex where the pulse energy is maximum, and also in the immediate vicinity of the vortex center. However, in many applications, the ring of maximum energy is not of primary interest, but the one where the peak intensity of the pulse is maximum, which is particularly true in nonlinear optics applications such as experiments with ultrafast vortices that excite high harmonics and attosecond pulses that also carry OAM. In this paper, the effects of the OAM-temporal coupling on the ring of maximum pulse peak intensity, which do not always coincide with the ring of maximum pulse energy, are described. We find that there is an upper limit to the magnitude of the topological charge that an ultrafast vortex with a prescribed pulse shape in its most intense ring can carry, and vice versa, a lower limit to the pulse duration in the most intense ring for a given magnitude of the topological charge. These limits imply that, with a given laser source spectrum, the duration of the synthesized ultrafast vortex increases with the magnitude of the topological charge. Explicit analytical expressions are given for the ultrafast vortices that contain these OAM-temporal couplings effects, which may be of interest in various applications, in particular in the study of their propagation and interaction with matter.

Keywords: ultrafast vortices; orbital angular momentum; pulses

1. Introduction

Although continuous light can transport vortices with an arbitrarily high magnitude of the topological charge \( l \), and therefore an arbitrarily high orbital angular momentum (OAM) (see, for example, [1,2]), pulsed light cannot [3,4]. This is a consequence of the coupling between the OAM and the temporal degrees of freedom in the pulsed vortices, as described also in [5,6].

Theoretically, this coupling affects pulsed vortices of any duration and topological charge, but it is only with the advances of the current technology in the generation of increasingly shorter vortices [7–15] of high quality (without angular and topological charge dispersions), at high powers, and approaching the single-cycle regime, and with the use of these ultrafast vortices in strong-field light–matter interactions for the generation of high harmonics and attosecond pulses with increasingly high OAM [16–20], that the effects of this coupling will be observable. They will then have an impact on ultrafast vortex applications such as classical and quantum information and entanglement [21–24], transfer of OAM to matter [25], materials processing [26], or nanosurgery [27]. Understanding the effects of OAM-temporal coupling is also fundamental when using the chirality of these vortices to probe the chiral properties of matter on ultrashort time scales [28,29].

Spatiotemporal couplings in ultrashort, pulsed light beams have been known for a long time (see, e.g., [30]), particularly in ultrashort, fundamental Gaussian beams in their various forms [31,32,33–35].
The OAM-temporal coupling is a particular spatiotemporal coupling, namely an azimuthal-temporal coupling, which seems to be rather more pronounced than the radial-temporal coupling in ultrashort Gaussian beams. In [3–6], the effects of OAM-temporal coupling are described for the first time in ultrafast vortices shaped as the pulsed Laguerre–Gauss (LG) beams commonly used in the experiments, as well as shaped as pulsed Bessel beams or diffraction-free X-waves. These effects are reviewed in [36], where it turns out that the OAM-temporal coupling manifests itself with different effects in different parts of the vortex. With a given broadband laser source, the carrier frequency experiences an important blue shift with respect to the laser mean frequency, \( \bar{\omega} \) (defined in the standard way [37] as the centroid of the spectral density), in the immediate vicinity of the vortex center, which is accompanied by an increment the number of oscillations, and these two effects are more pronounced as the magnitude of the topological charge is larger [4,36]. On the contrary, in the ring surrounding the vortex where the pulse energy is maximum, or “bright” ring for a time-integrating detector, the mean or carrier frequency is approximately the same as that of the laser source regardless of the topological charge, and the number of oscillations and duration of the pulse, \( \Delta t \) (defined by means of the central second-order moment of the pulse intensity), are larger than those obtainable with the available spectral bandwidth of the laser source, increasing with the magnitude of the topological charge in such a way that the duration is always above the minimum possible duration, \( \sqrt{|l|}/\bar{\omega} \), of an ultrafast vortex of charge \( l \) [3,5,36].

In this paper, we point out that the ring of the ultrafast vortex where the pulse energy is maximum is not always of primary interest. High pulse energy may result from long duration with relatively low peak intensity, while as short and as intense as possible pulses (resulting probably in lower energy) are typical demands in ultrafast nonlinear optics experiments, particularly in strong-field laser–matter interactions [16–20]. In these situations, minimum duration with a given laser source spectrum is obtained with uniform spectral phases, i.e., with transform-limited or unchirped pulses. This is why we limit our considerations to transform-limited pulses, and describe the effects of OAM-temporal coupling in the ring of the ultrafast vortex where the pulse peak intensity is maximum, which generally does not coincide with the ring of maximum pulse energy, as demonstrated below.

It turns out that the effects of OAM on the transform-limited pulse with maximum peak intensity are directly related to the properties of its symmetric temporal lobe around the pulse peak, which is, on the other hand, the only portion of the pulse of interest in nonlinear optics applications. Specifically, the involved magnitudes are the instantaneous frequency (derivative of the phase of the pulse with respect to time) at the instant of pulse peak, or “central” frequency, \( \omega_c \), and the duration of the central lobe around the peak of the pulse, defined by the concavity of the lobe, and which we call “central” duration, \( \Delta t_c \). As seen below, these two magnitudes are trivially determined by means of a measurement of the pulse frequency spectrum.

We then find qualitatively similar OAM-coupling effects in the most intense ring to those occurring in the most energetic ring. The central frequency of the pulse in the most intense ring is the same as the central frequency of the ultrafast laser source, and therefore independent of the imprinted topological charge. The duration of the central lobe is larger than that obtainable from the laser source without OAM, and increases with the magnitude of the topological charge. This effect is a result of the existence of a lower bound, \( \sqrt{|l|}/\bar{\omega}_c \), to the duration of the central temporal lobe that is satisfied by all existing ultrafast vortices in their most intense ring. If, as in many situations, the ring of maximum pulse energy coincides with the ring of maximum pulse peak intensity, then the pulse temporal shape in this ring is always such that the two inequalities, \( \Delta t > \sqrt{|l|}/\bar{\omega} \) for the pulse duration as a whole, and \( \Delta t_c > \sqrt{|l|}/\bar{\omega}_c \) for the duration of the central temporal lobe, are satisfied.

2. Ultrafast Vortices and Previous Results

We express an ultrafast vortex of topological charge \( l \), or \( l \) units of orbital angular momentum, as the superposition

\[
E(r, \phi, z, t') = \frac{1}{\pi} \int_0^\infty \hat{E}_\omega(r, \phi, z)e^{-i\omega t'}d\omega,
\]  

(1)
of monochromatic LG beams
\[ \hat{E}_\omega(r, \phi, z) = \hat{a}_\omega D(z, \phi) \left[ \frac{\sqrt{2r}}{s_\omega(z)} \right] |l| e^{i\omega z} e^{i\phi}, \]
(2)
of the same topological charge \( l \), zero radial order, weights \( \hat{a}_\omega \), and different angular frequencies \( \omega \). In the above expressions \( (r, \phi, z) \) are cylindrical coordinates, \( c \) is the velocity of light in vacuum, and \( t' = t - z/c \) is the local time for a plane pulse. The factor \( D(z, \phi) = e^{-i|l|\tan^{-1}(z/z_R)} e^{i\phi} / \left[ 1 + (z/z_R)^2 \right]^{1/2} \), where \( z_R \) is the Rayleigh distance, accounts for the azimuthal phase variation, and the \( z \)-dependent amplitude attenuation and Gouy’s phase shift associated with diffraction. The complex beam parameter is \( q(z) = z - i z_R \), which is usually written in the form
\[ \frac{1}{q(z)} = \frac{1}{R(z)} + i \frac{2c}{\omega s_\omega(z)}, \]
where
\[ s_\omega(z) = s_\omega \sqrt{1 + \left( \frac{z}{z_R} \right)^2}, \quad s_\omega = \sqrt{\frac{2z_R c}{\omega}}, \]
are the width and waist width, respectively, of the fundamental \( (l = 0) \) Gaussian beam, and \( 1/R(z) = z/(z^2 + z_R^2) \) is the curvature of the wave fronts of Gaussian and LG beams. Being limited to positive frequencies, the optical field \( E \) in Equation (1) is the analytical signal complex representation of the real field \( \text{Re} E \) [38]. As in preceding works [3,5,36], the Rayleigh distance \( z_R \) is assumed to be independent of frequency, i.e., we adopt the so-called isodiffracting model [33,34,39,40] because it is the only situation in which the pulse temporal shape does not experience any change during propagation (except for the global complex amplitude \( D \)) no matter how high \( l \) is, as shown recently [3] and as desired for applications. This choice is also a way to isolate the pure effects of OAM on temporal shape, i.e., the OAM-temporal coupling, and thus distinguish it from propagation effects on pulse shape in models other than the isodiffracting model, and whose study is left to further research.

For a simpler analysis, we introduce the normalized radial coordinate \( \rho = r / \sqrt{2z_R c[1 + (z/z_R)^2]} \). A constant value of \( \rho \) represents a revolution hyperboloid, or caustic surface, expanding as the monochromatic LG beams do. Equation (1) with Equation (2) now reads
\[ E(\rho, \tau) = \frac{1}{\pi} \int_0^\infty \hat{E}_\omega(\rho) e^{-i\omega \tau} d\omega, \]
(5)
where
\[ \hat{E}_\omega(\rho) = \hat{a}_\omega D \left( \sqrt{2\rho} \right) |l| e^{i\phi} e^{-i\rho^2}, \]
(6)
and where \( \tau = t' - r^2 / 2cR(z) \) is the local time for the ultrafast vortex. The pulse front \( \tau = 0 \) determines the time of arrival of the pulse at each point of space, and is the same family of spherical surfaces (in the paraxial approximation) as the phase fronts of the superposed isodiffracting LG beams. According to Equations (5) and (6), the pulse temporal shape depends on the particular caustic surface \( \rho \), but not on propagation distance \( z \).

In [3], the caustic surface \( \rho_F \) where the time-integrated intensity, energy density, or fluence, \( F(\rho) = \int_0^\tau (\text{Re} E)^2 d\tau = (1/2) \int_0^\infty |E|^2 d\tau = (1/\pi) \int_0^\infty |\hat{E}_\omega|^2 d\omega \) is maximum, i.e., the bright ring as recorded by a time-integrating detector, is considered the most relevant caustic surface, and the effects of OAM on pulse temporal shape on that caustic surface are described. The caustic surface of maximum fluence was shown to be determined by relation \( \rho_F^2 = |l| / 2\omega(\rho_F) \), where
\[ \omega(\rho) = \frac{\int_0^\infty |\hat{E}_\omega(\rho)|^2 \omega d\omega}{\int_0^\infty |\hat{E}_\omega(\rho)|^2 d\omega} \]
(7)
defines, as in [37], the mean frequency of the pulse on the caustic \( \rho \). On this caustic surface the pulse temporal bandwidth and the topological charge were shown to be restricted by inequality

\[
\Delta \omega(\rho_F) / \bar{\omega}(\rho_F) < 2 / \sqrt{|l|},
\]

where

\[
\Delta \omega^2(\rho) = \frac{\int_0^\infty |\hat{E}_\omega(\rho)|^2 (\omega - \bar{\omega}(\rho))^2 d\omega}{\int_0^\infty |\hat{E}_\omega(\rho)|^2 d\omega} \tag{8}
\]

is the so-called Gaussian-equivalent half-bandwidth on a caustic \( \rho \) (yielding the \( 1/e^2 \)-decay half-bandwidth for a Gaussian-like spectral density \( |\hat{E}_\omega(\rho)|^2 \)). Defining correspondingly the Gaussian-equivalent half-duration

\[
\Delta t^2(\rho) = \frac{\int_{-\infty}^\infty |E(\rho, \tau)|^2 (\tau - \bar{\tau}(\rho))^2 d\tau}{\int_{-\infty}^\infty |E(\rho, \tau)|^2 d\tau} \tag{9}
\]

where

\[
\bar{\tau}(\rho) = \frac{\int_{-\infty}^\infty |\hat{E}(\rho, \tau)|^2 d\tau}{\int_{-\infty}^\infty |\hat{E}_\omega(\rho)|^2 d\omega} \tag{10}
\]

and on account that \( \Delta t(\rho_F) \Delta \omega(\rho_F) \geq 2 \) for any pulse shape, it follows that the pulse duration on the bright ring of any ultrafast vortex always satisfies \( \Delta t(\rho_F) \geq \sqrt{|l|} / \bar{\omega}(\rho_F) \), which settles a lower bound to the duration of an ultrafast vortex with \( l \) units of OAM. It has been shown later [5,36] that the mean frequency on \( \rho_F \) is approximately equal to the mean frequency of \( \hat{a}_\omega \), which is directly related to the ultrashort laser source spectrum, i.e., \( \bar{\omega}(\rho_F) \approx \bar{\omega} \), where

\[
\bar{\omega} = \frac{\int_0^\infty |\hat{a}_\omega|^2 d\omega}{\int_0^\infty |\hat{a}_\omega|^2 d\omega}. \tag{11}
\]

With \( \bar{\omega}(\rho_F) \approx \bar{\omega} \) independent of the topological charge \( l \), the approximate lower bound \( \Delta t(\rho_F) > \sqrt{|l|} / \bar{\omega} \) with the right hand side growing monotonously with \( |l| \) implies that the duration in the bright ring of the synthesized ultrafast vortex must necessarily increase with \( |l| \) [5], if the laser source spectrum \( \hat{a}_\omega \) is fixed. These effects of the coupling between the OAM and the temporal degrees of freedom at the most energetic ring have been recently reviewed in [36], where they are also demonstrated to be substantially the same for other types of ultrafast vortices, as OAM-carrying nondiffracting X-waves [4,6,36].

### 3. OAM-Temporal Couplings in the Most Intense Ring of Transform-Limited Ultrafast Vortices

We point out here that the pulse of maximum energy is not always the ring of primary interest. In many applications, particularly in nonlinear optics experiments such as the generation of high harmonics and attosecond pulses excited by ultrafast vortices, the outcome of the experiment is primarily determined by the intensity, and possibly the carrier-envelope phase. Figure 1a,b illustrates that the caustic surface where the pulse has maximum energy and the caustic surface where the pulse has maximum peak intensity are generally different, and that the pulse shapes on these two caustic surfaces, including their mean frequencies and durations, are generally different.
Figure 1. (a) Radial profile of fluence, $F$, and peak intensity, $|E(\rho,0)|^2$, of the ultrafast vortex with topological charge $l = 27$ and spectrum $\hat{a}_\omega = \omega^{\nu} e^{-\omega^{2}/2\nu}$ of mean frequency $\bar{\omega} = 2.5$ $fs^{-1}$, normalized to their respective maxima on the respective caustic surfaces $\rho_F = 2.32$ $fs^{-1/2}$ and $\rho_E = 1.90$ $fs^{-1/2}$.

(b) Respective pulse shapes (real field and amplitudes) on these two caustic surfaces, oscillating at different carrier frequencies, $\bar{\omega}(\rho_F) = 2.5$ $fs^{-1} \simeq \bar{\omega}$ and $\bar{\omega}(\rho_E) = 3.616$ $fs^{-1}$, and with different durations. For a better comparison, the four curves are normalized to the peak amplitude of the pulse at $\rho_E$.

When the primary interest is maximum peak intensity, transform-limited pulses are desired because they have minimum duration with the available bandwidth, and then maximum intensity. The frequency spectrum $\hat{E}_\omega = \hat{b}_\omega D(\sqrt{2} \rho)^{|l|} \omega^{|l|/2} e^{-\rho^2 \omega}$ at every point of the ultrafast vortex has uniform spectral phases, and then the pulse $E(\rho, \tau)$ is transform-limited, if the laser source spectrum $\hat{a}_\omega$ has uniform phases, e.g., $\hat{a}_\omega = \hat{b}_\omega e^{-i\Phi}$, and then

$$\hat{E}_\omega = \hat{b}_\omega e^{-i\Phi} D(\sqrt{2} \rho)^{|l|} \omega^{|l|/2} e^{-\rho^2 \omega},$$

where $\hat{b}_\omega$ can be taken as real and non-negative, and $\Phi$ determines the carrier-envelope phase. Under these conditions, the pulse $E(\rho, \tau)$ consists of oscillations under an amplitude $|E(\rho, \tau)|$ that contains a symmetric central lobe about its peak value at the instant $\tau = 0$, whose value is

$$|E(\rho, \tau = 0)| = |D| \left( \sqrt{2} \rho \right)^{|l|} \frac{1}{\pi} \int_0^\infty \hat{b}_\omega \omega^{|l|/2} e^{-\rho^2 \omega} d\omega. \tag{13}$$

With $l \neq 0$, the peak amplitude in Equation (13) vanishes at $\rho = 0$ and for $\rho \to \infty$, so that there exists a particular caustic surface, say $\rho_E$, where the peak pulse amplitude is also maximum radially. The purpose of the following analysis is to describe the effects of OAM-temporal coupling on the caustic surface $\rho_E$ of ultrafast vortices.

3.1. Pulse Characterization

As a preliminary, we introduce a characterization of transform-limited pulses by means of two measurable parameters that will be seen to be directly involved in the OAM-temporal couplings. This characterization appears to be particularly relevant to nonlinear optics applications with these pulses because it describes quite accurately the shape of the main temporal lobe of amplitude about its maximum, and ignores the temporal parts of the pulse of low amplitude. First, we consider the instantaneous frequency at the time $\tau = 0$ of maximum amplitude, defined as

$$\omega_c(\rho) = \frac{d \arg E(\rho, \tau)}{d\tau} \bigg|_{\tau = 0}, \tag{14}$$
which is called the central frequency to distinguish it from $\bar{\omega}$, since their values are generally different. Writing $\text{arg} E = \tan^{-1}(\text{Im} E / \text{Re} E)$ with $\text{Re} E = (E + E^*)/2$ and $\text{Im} E = (E - E^*)/2i$, and using Equation (5) and its derivative with respect to time, one arrives at the alternate expression

$$\bar{\omega}_c(\rho) = \frac{\int_0^\infty \hat{E}_\omega(\rho) \omega d\omega}{\int_0^\infty \hat{E}_\omega(\rho) d\omega} = \frac{\int_0^\infty |\hat{E}_\omega(\rho)| \omega d\omega}{\int_0^\infty |\hat{E}_\omega(\rho)| d\omega}. \quad (15)$$

The introduction of the absolute values in the last step does not alter the result with the spectral phases in Equation (12) independent of frequency. In this way, the value of $\bar{\omega}_c$ is easily obtainable from the square root of the spectral density $|\hat{E}_\omega|^2$ measured by a spectrometer. Second, a measure of the width of the central lobe of the pulse based on its concavity is provided by the expression

$$\Delta t^2_c(\rho) = -2 \left( \frac{d^2\rho}{d\rho^2} \right)|_{\bar{\omega}_c(\rho)}^{-\frac{1}{2}}, \quad (16)$$

yielding the $1/e$-decay half-duration for a Gaussian-shaped amplitude. For other pulse shapes, Equation (16) provides the duration of a Gaussian pulse with the same concavity of the maximum at $\tau = 0$, and hence is called the central duration. Using the equivalent expression $\Delta t^2_c = -4|E|^2/(d^2|E|^2/d\tau^2)_{\tau=0}$, writing $|E|^2 = E E^*$, using Equation (5) and its derivatives with respect to time at $\tau = 0$, one easily arrives at the alternate expression

$$\Delta t^2_c(\rho) = 4/\Delta \omega^2_c(\rho), \quad (17)$$

where

$$\Delta \omega^2_c(\rho) = 2 \frac{\int_0^\infty \hat{E}_\omega(\rho) (\omega - \bar{\omega}_c(\rho))^2 d\omega}{\int_0^\infty \hat{E}_\omega(\rho) d\omega} = \frac{2}{\int_0^\infty \hat{E}_\omega(\rho) |\omega - \bar{\omega}_c(\rho)|^2 d\omega} \quad (18)$$

defines a spectral bandwidth based on the spectral amplitude instead of the spectral intensity that yields, again, the $1/e$-decay half-bandwidth for a Gaussian-like spectral amplitude $|\hat{E}_\omega(\rho)|$, and therefore coincides with $\Delta \omega(\rho)$ for a Gaussian spectrum. The measures $\Delta \omega$ and $\Delta \omega_c$ are however different for other spectral shapes; in particular, $\Delta \omega_c$ overweights widespread frequencies far from the central frequency of the spectrum compared to $\Delta \omega$. The two parameters $\omega_c$ and $\Delta t_c$ allow reconstructing the shape of the central portion the pulse very reliably. Figure 2 shows two ultrashort pulse shapes and their approximation $e^{-i\tau^2/\Delta t^2_c} e^{-i(\omega_c \tau + \Phi)}$ to its central lobe of high amplitude based on the central frequency and duration.

![Figure 2](image-url) Figure 2. The pulse $E = [1/(1 + \tau^2/T^2)]e^{-i(\omega_0 \tau + \Phi)}$ with $\omega_0 = 2.5 \text{ rad fs}^{-1}$, $\Phi = \pi/6$, and (a) $T = 1$ fs and (b) $T = 4$ fs (solid black curves), their amplitudes $|E|$ (dashed black curves), and their approximations $E_c = e^{-i\tau^2/\Delta t^2_c} e^{-i(\omega_c \tau + \Phi)}$ (gray solid curves) and $|E_c|$ (dashed gray curves) based on the central frequency $\omega_c = 2.5 \text{ fs}^{-1}$ and the central width $\Delta t_c = T$. 
3.2. Lower Bound to the Ultrafast Vortex Duration in Its Most Intense Ring

With the above pulse properties in mind, let us locate the caustic surface $\rho_E$ of the ultrafast vortex where the peak pulse amplitude is maximum. After some calculations, the derivative of the peak amplitude in Equation (13) with respect to $\rho$ is found to be

$$\frac{d|E(\rho,0)|}{d\rho} = \frac{|E(\rho,0)|}{\rho} \left[ |l| - 2\rho^2 \omega_c(\rho) \right].$$  \hfill (19)

The fluence will then be maximum or minimum at the caustic surface determined by

$$\rho_E^2 = \frac{|l|}{2\omega_c(\rho_E)},$$  \hfill (20)

which generally differs from the caustic surface $\rho_E^2 = |l|/2\bar{\omega}(\rho_F)$ of maximum fluence [3]. Evaluation of the second derivative of $|E(\rho,0)|$ with respect to $\rho$ leads to an involved expression, which however at $\rho_E$ simplifies to

$$\frac{d^2|E(\rho,0)|}{d\rho^2} \bigg|_{\rho_E} = -2\bar{\omega}_E(\rho_E)|E(\rho_E,0)| \left[ 2 - \frac{|l|}{|\omega^2(\rho_E)|} \right].$$  \hfill (21)

The condition of maximum in Equation (21) allows us to conclude that the bandwidth $\Delta\omega_c$ on the caustic surface of maximum intensity and the topological charge of ultrafast vortices always satisfy the inequality

$$\frac{\Delta\omega_c(\rho_E)}{\omega_c(\rho_E)} < \frac{2}{\sqrt{|l|}},$$  \hfill (22)

i.e., the relative bandwidth on the most intense ring cannot exceed a maximum value determined by the topological charge of the vortex. The inequality in Equation (22) is formally equal as $\Delta\omega(\rho_F)/\bar{\omega}(\rho_F) < 2/\sqrt{|l|}$, but these two inequalities refer to different caustic surfaces and the quantities involved are different. The above inequality implies a lower bound to the duration of the central lobe of an ultrafast vortex at its most intense ring. From Equation (17), the inequality in Equation (22) transforms into

$$\Delta t_c(\rho_E) > \frac{\sqrt{|l|}}{\omega_c(\rho_E)}.$$

This inequality imposes a lower bound to the central duration relative to the central period $2\pi/\omega_c(\rho_E)$, and therefore a lower bound to the number of oscillations of the pulse in the most intense ring of the ultrafast vortex. If the number of oscillations $N$ is measured as the full width at half maximum of intensity of the central lobe, $\sqrt{2\ln 2}\Delta t_c(\rho_E)$, over the central period, the inequality in Equation (23) leads to inequality $N > \sqrt{2/\pi} \sqrt{|l|}/2\pi$ for the number of oscillations. Since the lower bound in the right hand side increases monotonically with $|l|$, the number of oscillations in the central lobe of the pulse in the most intense ring of the ultrafast vortices synthesized with the same source spectrum $\bar{a}_\omega$ increases if the imprinted topological charge is increased. In practice, as seen below, the central frequency $\omega_c(\rho_E)$ of the pulse in the most intense ring is substantially determined by the available frequencies in the superposition, i.e., by $\bar{a}_\omega$, indeed $\omega_c(\rho_E) \approx \bar{\omega}_c$, with

$$\bar{\omega}_c = \frac{\int_0^\infty |\bar{a}_\omega|d\omega}{\int_0^\infty |\bar{a}_\omega|d\omega},$$  \hfill (24)

With $\omega_c(\rho_E) \approx \bar{\omega}_c$, independent of the topological charge, the inequality in Equation (23) imposes directly a lower bound $\sqrt{|l|}/\bar{\omega}_c$ to the duration of the main lobe, and implies an increase of this duration with the magnitude of the topological charge.
4. Ultrafast Vortices with Prescribed Pulse Shape in the Most Intense Ring

As a verification, suppose we need the expression for an ultrafast vortex of certain transform-limited temporal shape,

$$P(\tau) = \frac{1}{\pi} \int_0^\infty \hat{P}_\omega e^{-i\omega \tau} d\omega,$$

(25)
in the most intense ring. The pulse $P(\tau)$ is characterized by certain central frequency $\bar{\omega}_c$ and duration $\Delta \tau_c$. Equating Equations (5) and (6) evaluated at $\rho^2 = |l|/2\bar{\omega}_c(\rho_E) = |l|/2\bar{\omega}_c$ to $DP(\tau)$, the needed laser source spectrum must be $\hat{a}_\omega = \hat{P}_\omega \omega^{-|l|/2\bar{\omega}_c^2(\sqrt{2}\rho_E)^{-|l|}}$. Introducing this expression of $\hat{a}_\omega$ in Equation (6) for generic $\rho$, the result in Equation (5), and performing the integral, we obtain the expression

$$E(\rho, \tau) = D \left( \frac{\rho}{\rho_E} \right)^{|l|} P \left( \tau - i\rho^2 + i\rho_E^2 \right),$$

(26)
of the ultrafast vortex of pulse shape $P(\tau)$ at $\rho_E$. Coming back to the physical radius $r$, the caustic of maximum peak intensity reads $r_E(z) = \sqrt{|l|/2 s_{\bar{\omega}_c}(z)}$, where $s_{\bar{\omega}_c}$ is given by Equation (4) evaluated at $\bar{\omega}_c$, and the ultrafast vortex reads

$$E = D \left( \sqrt{\frac{2}{|l|}} \frac{r}{s_{\bar{\omega}_c}(z)} \right)^{|l|} P \left( \tau' - r^2/(2cq(z)) + i \frac{|l|}{2\bar{\omega}_c} \right).$$

(27)

With given $P(\tau)$, and hence given $\bar{\omega}_c$ and $\Delta \tau_c$, Equation (27) represents indeed an ultrafast vortex with pulse shape $P(\tau)$ in its most intense ring at $r_E(z) = \sqrt{|l|/2 s_{\bar{\omega}_c}(z)}$ only if $|l| < \bar{\omega}_c^2 \Delta \tau_c^2$; otherwise, the intensity takes a relative minimum at $r_E(z) = \sqrt{|l|/2 s_{\bar{\omega}_c}(z)}$, with a maximum at another radius with different pulse shape such that the inequality in Equation (23) is satisfied, or possibly a singular expression, as in the following example.

We consider the transform-limited pulses

$$P(\tau) = \left( \frac{-i\beta}{\bar{\omega}_c \tau - i\beta} \right)^\beta e^{-i\phi},$$

(28)
where $\bar{\omega}_c$ is just the central frequency, and the parameter $\beta \geq 1$ determines the central bandwidth $\Delta \omega_c = \sqrt{2|l|/\beta} \bar{\omega}_c$ and the duration $\Delta \tau_c = \sqrt{2|l|/\bar{\omega}_c}$. For $\beta \gg 1$, $P(\tau)$ approaches the Gaussian pulse $\exp(-\tau^2/\Delta \tau_c^2) e^{-i(\bar{\omega}_c \tau + \phi)}$ based on its central parameters not only in its central portion but at any time because $P(\tau)$ approaches a Gaussian-enveloped pulse, as in the example of Figure 3a showing a near infrared, single-cycle pulse ($\bar{\omega}_c = 2.417$ rad/fs, $\beta = 14.24$). Condition $|l| < \bar{\omega}_c^2 \Delta \tau_c^2$ for the existence of the ultrafast vortex with this pulse shape in its most intense ring reads $|l| < 2\beta$. Indeed, Equation (27) with Equation (28) yields

$$E = D \left( \sqrt{\frac{2}{|l|}} \frac{r}{s_{\bar{\omega}_c}(z)} \right)^{|l|} \left[ \frac{-i\beta}{\bar{\omega}_c \left( \tau' - \frac{r^2}{2cq(z)} \right) + i \left( \frac{|l|}{2\bar{\omega}_c} \beta \right) \phi} \right] e^{-i\phi}.$$

(29)
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Figure 3. (a) The real part (black solid) and amplitude (black dashed) of the pulse $P$ in Equation (28) with $\omega_c = 2.417$ rad/fs (780 nm wave length), $\beta = 14.24$, and $\Phi = \pi/2$, containing a single oscillation within its full width at half maximum in intensity, and its approximation $P_c = e^{-\tau^2/\Delta t_c^2}e^{-i(\omega_c \tau + \Phi)}$, with $\Delta t_c = 2.08$ fs (gray curves). (b) Peak amplitudes of the ultrafast vortices of Rayleigh distance $z_R = 1$ m with the pulse shape $P(\tau)$ at $r_F(z) = \sqrt{|l|/2} s_{\omega_c}(z)$ as functions of radius as given by Equation (30) at $z = 0$ and for increasing values of $l$, featuring maxima at $r_F(0) = \sqrt{|l|/2} s_{\omega_c}(0)$ for $|l| < 2\beta$ and minima at $r_F(0) = \sqrt{|l|/2} s_{\omega_c}(0)$ for $|l| > 2\beta$, with a singularity at a smaller radius, and thus lacking physical meaning.

Its peak amplitude at $\tau = 0 \ [t' = r^2/2cR(z)]$,

$$|E(\tau = 0)| = D \left( \sqrt{\frac{2}{|l|}} \frac{r}{s_{\omega_c}(z)} \right)^{|l|} \left( \frac{\beta}{\sqrt{r^2/s_{\omega_c}(z)^2 - |l|^2/\beta^2}} \right)^{\beta} \tag{30}$$

as a function of $r$ is depicted in Figure 3b for increasing values of $|l|$, and is characterized by a maximum at the corresponding radii $r_F(z) = \sqrt{|l|/2} s_{\omega_c}(z)$ if $|l| \leq \omega_c^2 \Delta t_c^2$ (i.e., if $|l| < 2\beta = 28.48$), but by a minimum at $r_F(z) = \sqrt{|l|/2} s_{\omega_c}(z)$ if $|l| \geq \omega_c^2 \Delta t_c^2$ (i.e., if $|l| > 2\beta = 28.48$) because the peak amplitude in Equation (30) has a singularity at a radius smaller than $r_F(z)$, which makes Equation (29) physically invalid. Thus, an ultrafast vortex whose pulse shape in its most intense ring is a single-cycle pulse can carry only up to 28 units of OAM.

4.1. Ultrafast Vortices with Power-Exponential Laguerre–Gauss Spectrum

In practice, one has a spectrum $\delta_\omega$ that is substantially determined by the laser source and is therefore independent of the topological charge $l$, and with this spectrum one may wish to synthesize an ultrafast vortex of certain charge $l$. The above OAM-temporal coupling effects imply that the temporal properties of the main lobe of the pulse in the most intense ring of the ultrafast vortices synthesized with the same spectrum $\delta_\omega$ change with $l$. This is particularly true if the bandwidth of $\delta_\omega$ is very large and/or the topological charge high.

As a sufficiently flexible model, we take the power-exponential spectrum

$$\delta_\omega = \frac{\pi}{\Gamma(\beta)} \left( \frac{\beta}{\omega_c} \right)^{-1} \omega_c^\beta e^{-\omega_c^\beta} e^{-i\Phi}, \tag{31}$$

corresponding in time domain to the same pulse $a(\tau) = [-i\beta/(\omega_c \tau - i\beta)]^{\beta} e^{-i\Phi}$ as $P(\tau)$ in Equation (28) of arbitrary central frequency $\omega_c$ and central duration $\Delta t_c = \sqrt{2\beta}/\omega_c$. Integral in
Equation (1) with the LG beams in Equation (1) and with the power-exponential spectrum in Equation (31) can be carried out to yield the closed-form expression

\[
E = D \left( \sqrt{\frac{2}{|l|}} \frac{r}{s \bar{\omega}_c(z)} \right)^{|l|} \left[ \frac{-i \left( \frac{\beta + |l|}{\tau} \right)}{\bar{\omega}_c \left( t' - \frac{r^2}{2z_0(z)} \right) - i \beta} \right]^{\beta + \frac{\gamma}{2}} e^{-i \Phi}
\]

which is, this time, nonsingular and localized for any value of \( l \). A multiplicative factor has been introduced in Equation (32) to adjust to unity the peak amplitude at \( rE(0) = \sqrt{|l|/2} s \bar{\omega}_c \) at the waist \( z = 0 \). The peak amplitude at any point \((r, z)\) is given by

\[
|E(\tau = 0)| = D \left( \sqrt{\frac{2}{|l|}} \frac{r}{s \bar{\omega}_c(z)} \right)^{|l|} \left( \frac{\beta + \frac{|l|}{\tau}}{\bar{\omega}_c + \frac{r^2}{2z_0(z)}} \right)^{\beta + \frac{|l|}{2}}
\]

which takes a maximum value on the caustic surface \( rE(z) = \sqrt{|l|/2} s \bar{\omega}_c(z) \). On this caustic surface, the pulse shape is

\[
E(\tau) = D \left[ \frac{-i \left( \frac{\beta + |l|}{\tau} \right)}{\bar{\omega}_c \tau - i \left( \frac{\beta + |l|}{\tau} \right)} \right]^{\beta + \frac{|l|}{2}} e^{-i \Phi},
\]

which is the same as \( a(\tau) \) but with \( \beta \) replaced with \( \beta + |l|/2 \). Thus, for the fundamental Gaussian beam \( (l = 0) \), i.e., the fundamental isodiffracting pulsed Gaussian beam [33,34], the pulse shape at \( r = 0 \) is just \( a(\tau) \) of the central frequency \( \bar{\omega}_c \) and duration \( \Delta t_c = \sqrt{2\beta}/\bar{\omega}_c \), as determined by the full source spectrum \( \hat{a}_\omega \). For \( |l| \neq 0 \), the pulse temporal shape in the most intense ring maintains the frequency \( \bar{\omega}_c(rE) = \bar{\omega}_c \) of the source spectrum regardless of the value of \( l \), but the duration increases with the magnitude of the topological charge as

\[
\Delta t_c(rE) = \sqrt{\frac{2\beta + |l|}{\bar{\omega}_c}};
\]

in particular, the pulse adapts itself so that its duration is always above the lower bound \( \sqrt{|l|/\bar{\omega}_c(rE)} = \sqrt{|l|/\bar{\omega}_c} \). Figure 4 illustrates the adaptation of pulse shape on the most intense caustic surface to the value of the topological charge. With the ultra-broadband spectrum of Figure 4a of central frequency in the near infrared, the half-cycle pulse represented by blue curves in Figure 4b–d could be built with \( l = 0 \) at \( r = 0 \). At the maxima of the radial distribution of peak intensity depicted in Figure 4e for several values of \( |l| \), the pulses represented in Figure 4b–d as black curves are seen to increase their duration with increasing \( |l| \). As a summary of this OAM-temporal coupling effect, the black curves in Figure 4f represent the pulse duration as a function of the topological charge for increasing bandwidths of \( \hat{a}_\omega \) that correspond to laser pulses with increasing number \( N \) of cycles. Irrespective of the source bandwidth and associated number of cycles, the duration in the most intense ring of the produced ultrafast vortex is always above \( \sqrt{|l|/\bar{\omega}_c} \), the adaptation being more pronounced as the number of cycles is smaller and vanishing in the monochromatic limit.
Figure 4. (a) Power-exponential spectrum $\hat{a}_\omega$ in Equation (31) with $\bar{\omega}_c = 2.417$ rad/fs and $\beta = 3.56$. (b–d) Real part and amplitude of the half-cycle ($N = 0.5$) pulse $a(\tau)$ in time domain (blue curves), and real part and amplitude of the pulse in the most intense ring of ultrafast vortices with the indicated values of $|l|$, as given by Equation (34). All pulse shapes are normalized to their peak values for a better comparison. (e) With $z_R = 10$ mm, peak intensity of the pulse as a function of radius at the waist $z = 0$ for the three values of $|l|$ with maxima at $r_E(0) = \sqrt{|l|/2s_{\omega_c}}$. (f) Central duration of vortices in their most intense ring as a function of the magnitude of the topological charge $|l|$, for power-exponential laser spectra with $\bar{\omega}_c = 2.417$ rad/fs, and with $\beta = 3.56$ ($N = 0.5$), $\beta = 14.24$ ($N = 1$), $\beta = 32.04$ ($N = 1.5$), $\beta = 56.95$ ($N = 2$), $\beta = 89.0$ ($N = 2.5$), and $\beta = 128.15$ ($N = 3$). The red curve is the lower limit to the pulse duration in the inequality in Equation (23).

5. Conclusions

We describe how the OAM-temporal coupling in ultrafast vortices affects the temporal properties of the vortex on the ring of maximum peak intensity, which is the ring of primary relevance in many nonlinear optics applications, particularly those involving strong-field light–matter interactions. In a few words, the ultrafast vortices synthesized with a given laser source spectrum increase their duration
if the imprinted topological charge is increased in order for the pulse duration on the most intense ring to always remain above a lower limit that increases monotonically with the magnitude of the topological charge. We provide analytical expressions of ultrafast vortices that incorporate these effects and can be used as a starting point for the study of their linear and nonlinear interaction with matter. Although we limit our considerations to ultrafast vortices in optics, the same OAM-temporal coupling effects affect ultrafast electron vortices [41] and transient acoustic vortices [42].

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**Abbreviations**

The following abbreviations are used in this manuscript:

- LG Laguerre–Gauss
- OAM Orbital Angular Momentum

**References**

1. Allen, L.; Beijersnergen, M.W.; Spreeuw, R.J.C.; Woerdman, J.P. Orbital angular momentum of light and the transformation of Laguerre-Gaussian laser modes. *Phys. Rev. A* 1992, 45, 8185–8189.
2. Yao, A.M.; Padgett, M.J. Orbital angular momentum: Origins, behavior and applications. *Adv. Photonics* 2011, 3, 161–204.
3. Porras, M.A. Upper Bound to the Orbital Angular Momentum Carried by an Ultrashort Pulse. *Phys. Rev. Lett.* 2019, 122, 123904.
4. Ornigotti, M.; Conti, C.; Szameit, A. Effect of Orbital Angular Momentum on Nondiffracting Ultrashort Optical Pulses. *Phys. Rev. Lett.* 2015, 115, 100401.
5. Porras, M.A. Effects of orbital angular momentum on few-cycle and sub-cycle pulse shapes: Coupling between the temporal and angular momentum degrees of freedom. *Opt. Lett.* 2019, 44, 2538–2541.
6. Ornigotti, M.; Conti, C.; Szameit, A. Universal form of the carrier frequency of scalar and vector paraxial X waves with orbital angular momentum and arbitrary frequency spectrum. *Phys. Rev. A* 2015, 92, 043801.
7. Bezuhanov, K.; Dreischuh, A.; Paulus, G.G.; Schätzel, M.G.; Walther, H. Vortices in femtosecond laser fields. *Opt. Lett.* 2004, 29, 1942–1944.
8. Mariyenko, I.G.; Strohaber, J.; Uiterwaal, C.J.G.J. Creation of optical vortices in femtosecond pulses. *Opt. Express* 2005, 13, 7599–7608.
9. Bezuhanov, K.; Dreischuh, A.; Paulus, G.; Schätzel, M.G.; Walther, H.; Neshev, D.; Krolikowski, W.; Kivshar, Y. Spatial phase dislocations in femtosecond laser pulses. *J. Opt. Soc. Am. B* 2006, 23, 26–35.
10. Zeylikovich, I.; Sztul, H.I.; Kartazaev, V.; Le, T.; Alfano, R.R. Ultrashort Laguerre-Gaussian pulses with angular and group velocity dispersion compensation. *Opt. Lett.* 2007, 32, 2025–2027.
11. Tokizane, Y.; Oka, K.; Morita, R. Supercontinuum optical vortex pulse generation without spatial or topological-charge dispersion *Opt. Express* 2009, 17, 14517–14525.
12. Shvedov, V.G.; Hnatovsky, C.; Krolikowski, W.; Rode, A.V. Efficient beam converter for the generation of high-power femtosecond vortices. *Opt. Lett.* 2010, 35, 2660–2662.
13. Richter, A.; Bock, M.; Jahn, J.; Gruenwald, R. Orbital angular momentum experiments with broadband few cycle pulses. *Proc. SPIE* 2010, 7613, 761308.
14. Yamane, K.; Toda, Y.; Morita, R. Ultrashort optical-vortex pulse generation in few-cycle regime. *Opt. Express* 2012, 20, 18986–18993.
15. Miranda, M.; Kotur, M.; Rudawski, P.; Guo, C.; Harth, A.; L’Huillier, A.; Arnold, C.L. Spatiotemporal characterization of ultrashort optical vortex pulses. *J. Mod. Opt.* 2017, 64, S1–S6.
16. Hermández-García, C.; Picón, A.; San Román, J.; Plaja, L. Attosecond Extreme Ultraviolet Vortices from High-Order Harmonic Generation. *Phys. Rev. Lett.* 2013, 111, 083602.
17. Gariepy, G.; Leach, J.; Kim, K.T.; Hammond, T.J.; Frumker, E.; Boyd, R.W.; Corkum, P.B. Creating High-Harmonic Beams with Controlled Orbital Angular Momentum. *Phys. Rev. Lett.* 2014, 113, 153901.
18. Rego, L.; San Román, J.; Picón, A.; Plaja, L.; Hernández-García, C. Nonperturbative Twist in the Generation of Extreme-Ultraviolet Vortex Beams. *Phys. Rev. Lett.* 2016, 117, 163202.

19. Turpin, A.; Rego, L.; Picón, A.; San Romá, J.C.; Hernández-García, C. Extreme ultraviolet fractional orbital angular momentum beams from high harmonic generation. *Sci. Rep.* 2017, 7, 43888.

20. Rego, L.; Domey, K.M.; Brooks, N.J.; Nguyen, Q.L.; Liao, C.-T.; San Román, J.; Couch, D.E.; Liu, A.; Pisanty, E.; Lewenstein, M.; et al. Generation of extreme-ultraviolet beams with time-varying orbital angular momentum. *Science* 2019, 364, 1253.

21. Gibson, G.; Courtial, J.; Padgett, M.J.; Vasnetsov, M.; Pasko, V.; Barnett, S.M.; Franke-Arnold, S. Free-space information transfer using light beams carrying orbital angular momentum. *Opt. Express* 2004, 12, 5448–5456.

22. Huang, H.; Xie, G.; Yan, Y.; Ahmed, N.; Ren, Y.; Yue, Y.; Rogawski, D.; Willner, M.J.; Erkmen, B.I.; Birmbaum, K.M.; et al. 100 Tbit/s free-space data link enabled by three-dimensional multiplexing of orbital angular momentum, polarization, and wavelength. *Opt. Lett.* 2014, 39, 197–200.

23. Mair, A.; Vaziri, V.; Weihs, G.; Zeilinger, A. Entanglement of the orbital angular momentum states of photons. *Nature* 2001, 412, 313–316.

24. Molina-Terriza, G.; Torres, J.P.; Torner, L.L. Management of the Angular Momentum of Light: Preparation of Photons in Multidimensional Vector States of Angular Momentum. *Phys. Rev. Lett.* 2002, 88, 013601.

25. He, H.; Friese, M.E.J.; Heckenberg, N.R.; Rubinsztein-Dunlop, H. Direct observation of transfer of angular-momentum to absorptive particles from a laser-beam with a phase singularity. *Phys. Rev. Lett.* 1995, 75, 826–829.

26. Omatsu, T.; Chuo, K.; Miyamoto, K.; Okida, M.; Nakamura, K.; Aoki, N.; Morita, R. Metal microneedle fabrication using twisted light with spin. *Opt. Express* 2010, 18, 17967–17973.

27. Jeffries, G.D.M.; Scott Edgar, J.; Zhao, Y.; Shelby, J.P.; Fong, C.; Chiu, D.T. Using Polarization-Shaped Optical Vortex Traps for Single-Cell Nanosurgery. *Nano Lett.* 2007, 7, 415–420.

28. Ayuso, D.; Neufeld, O.; Ordonez, A.F.; Decleva, P.; Lerner, G.; Cohen, O.; Ivanov, M.; Smirnova, O. Synthetic chiral light for efficient control of chiral light–matter interaction. *Nat. Photonics* 2019, 13, 866–871.

29. Silva, R.E.F.; Jiménez-Galán, A.; Amorim, B.; Smirnova, O.; Ivanov, M.Y. Topological strong-field physics on sub-laser-cycle timescale. *Nat. Photonics* 2019, 13, 849–854.

30. Akturk, S.; Gu, X.; Bowlan, P.; Trebino, R. Spatio-temporal couplings in ultrashort laser pulses. *J. Opt.* 2010, 12, 093001.

31. Sheppard, C.J.R.; Gan, X. Free-space propagation of femto-second light pulses. *Opt. Commun.* 1997, 133, 1–6.

32. Kaplan, A.E. Diffraction-induced transformation of near-cycle and subcycle pulses. *J. Opt. Soc. Am. B* 1998, 15, 951–956.

33. Porras, M.A. Ultrashort pulsed Gaussian light beams. *Phys. Rev. E* 1998, 58, 1086–1093.

34. Porras, M.A. Nonsinusoidal few-cycle pulsed light beams in free space. *J. Opt. Soc. Am. B* 1999, 16, 1468–1474.

35. Porras, M.A. Diffraction effects in few-cycle optical pulses. *Phys. Rev. E* 2002, 65, 026606.

36. Porras, M.A.; Conti, C. Couplings between the temporal and orbital angular momentum degrees of freedom in ultrafast vortices with propagation-invariant temporal shape. *arXiv* 2019, arXiv:1911.12225.

37. Brabec T.; Krausz, F. Nonlinear Optical Pulse Propagation in the Single-Cycle Regime. *Phys. Rev. Lett.* 1997, 78, 3282–3285.

38. Born M.; Wolf, E. *Principles of Optics*; Pergamon: Oxford, UK, 1975.

39. Porras, M.A.; Borghi, R.; Santarsiero, M. Few-optical-cycle Bessel-Gauss pulsed beams in free space. *Phys. Rev. E* 2000, 62, 5729.

40. Feng, S.; Winful, H.G. Higher-order transverse modes of ultrashort isodiffracting pulses. *Phys. Rev. E* 2001, 63, 046602.
41. McMorran, B.J.; Agrawall, A.; Anderson, I.M.; Herzing, A.A.; Lezec, H.J.; McClelland, J.J.; Unguris, J. Electron Vortex Beams with High Quanta of Orbital Angular Momentum. *Science* **2011**, *331*, 192–195.

42. Marzo, A.; Caleap, M.; Drinkwater, B.W. Acoustic Virtual Vortices with Tunable Orbital Angular Momentum for Trapping of Mie Particles. *Phys. Rev. Lett.* **2018**, *120*, 044301.