Observation of subluminal twisted light in vacuum: supplementary material

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Published 23 March 2016

This document provides supplementary information to “Observation of subluminal twisted light in vacuum,” <http://dx.doi.org/10.1364/optica.3.000351>.” © 2016 Optical Society of America

<http://dx.doi.org/10.1364/optica.3.000351.s001>

Experimental setup: The output of a 100 fs pulsed Ti:Sa laser operating with a repetition rate of 82 MHz and an average power of 300 mW at a central wavelength of 795 nm is split into two arms by means of a non-polarising beam splitter (BS). In the test arm, the incident beam is made to pass through a lens placed immediately prior to a kinoform displayed on a Pluto-HOLOEYE spatial light modulator (SLM), which reshapes the spatial distribution of the incoming pulsed beam into a desired HyGG_ℓ mode. In order to generate a pure LG_{p,ℓ} mode, one would have to use an intensity masking technique. The downside to this method is the power reduction in the beam that accompanies the masking strategy, a crucial consideration in a nonlinear optical experiment, as the efficiency of the SHG signal depends on the intensity of the pump beam. No lenses are used after the hologram, in order to avoid the introduction of additional artificial time delays. Since the different LG modes considered have intensity maxima at different radii, a lens placed in the optical path of the beam would introduce an additional time delay, which would depend on the lens thickness at the radius of the beam's intensity maximum. We note that one may be tempted to overcome this difficulty by making use of flat (Fresnel) lenses. However, this would result in a distortion of the pulse shape and spectrum [1].

The first diffracted order obtained from the reflection of the test beam off the SLM is selected using an iris placed at the Fourier plane of the lens. The test and reference pulses are respectively focused by means of $f_1 = 400$ mm and $f_2 = 500$ mm lenses into a $L = 500$ μm thick type-I β-barium borate (BBO) crystal, where they are spatially overlapped. The Rayleigh range, $z_R = 2$ cm ≈ 40 L, associated with the test beam is much longer than the crystal length. The cross-SHG signal is isolated from the two other SHG outputs and fundamental beams by means of an iris followed by an

interference filter. The power associated with this signal is measured over a period of one second using a Newport power meter with a silicon detector.

Cross-correlation: The second-order polarisation arising from the reference (LG_{0,0})-test (LG_{p,ℓ}) beam interaction is given by $P_{0,\ell}^{(2)}(t) = \epsilon_0 \chi^{(2)} \text{LG}_{0,0}(t) \text{LG}_{p,\ell}(t - \tau)$, where $\text{LG}_{0,\ell}(t)$ represents the time-dependent electric field associated with the LG_{0,ℓ} mode, τ indicates the time delay between the two pulses, and $\chi^{(2)}$ is the second-order nonlinear susceptibility of the BBO crystal. The intensity associated with the cross-beam SHG signal is then proportional to the product $|\text{LG}_{0,0}(t)|^2 |\text{LG}_{p,\ell}(t - \tau)|^2$. Hence, maximal cross-beam SHG will be observed when the delay is set to be zero, $\tau = 0$. However, due to the Gaussian temporal profile of the pulses, some non-collinear SHG will result even in the case of small non-zero time delays. As the time delay between both pulses increases, the non-collinear SHG signal decreases gradually, disappearing when τ significantly exceeds the pulse temporal duration.

Phase and Group Velocities

A surface of constant phase, also known as a cophasal surface, associated with a monochromatic electric field $E(\mathbf{r}, t) = A(\mathbf{r})e^{i(\omega t - \Phi(\mathbf{r}))}$, where $A(\mathbf{r})$ and $\Phi(\mathbf{r})$ respectively denote the amplitude and phase of the electric field, is given by [2]

$$\omega t - \Phi(\mathbf{r}) = \text{constant}, \quad (\text{S1})$$

whence

$$\delta(\omega t - \Phi(\mathbf{r})) = 0. \quad (\text{S2})$$

The phase velocity describes the rate at which a point of constant phase travels through space, i.e. $v_{\text{ph}} = \delta r_{\text{ph}} / \delta t$, and can be calculated by

$$\omega \delta t - \nabla \Phi(\mathbf{r}) \cdot \delta \mathbf{r} = 0. \quad (\text{S3})$$

Equation (S3) can be rearranged by introducing a unit vector $\hat{\mathbf{r}} = \delta \mathbf{r} / \delta r_{\text{ph}}$, yielding

$$\begin{aligned} v_{\text{ph}} &= \frac{\delta r_{\text{ph}}}{\delta t} \\ &= \frac{\omega}{\mathbf{r} \cdot \nabla \Phi(\mathbf{r})} = \frac{\omega}{|\nabla \Phi(\mathbf{r})|}, \end{aligned} \quad (\text{S4})$$

where we have used the fact that $\hat{\mathbf{r}} = \nabla \Phi(\mathbf{r}) / |\nabla \Phi(\mathbf{r})|$ for propagation of a wavefront.

For polychromatic fields, which in general may be expressed in the form $E(\mathbf{r}, t) = \int_{-\infty}^{\infty} \tilde{A}(\mathbf{r}, \omega) e^{i(\omega t - \Phi(\mathbf{r}, \omega))} d\omega$, a group velocity may be assigned to the pulse envelope. Here, $\tilde{A}(\mathbf{r}, \omega)$ is the angular frequency amplitude and represents the amplitude of the field in frequency space. The group velocity may be derived by expressing the electric field as a product of its carrier and envelope components, such that $E(\mathbf{r}, t) = A(\mathbf{r}, t) e^{i(\omega_0 t - \Phi(\mathbf{r}, \omega_0))}$, where ω_0 stands for the central angular frequency of the field. From these two expressions for the electric field, it can be seen that

$$A(\mathbf{r}, t) = \int_{-\infty}^{\infty} \tilde{A}(\mathbf{r}, \omega) e^{i\{(\omega - \omega_0)t - (\Phi(\mathbf{r}, \omega) - \Phi(\mathbf{r}, \omega_0))\}} d\omega. \quad (\text{S5})$$

The phase Φ_{envelope} of the field envelope therefore evolves according to $\Phi_{\text{envelope}} = (\omega - \omega_0)t - (\Phi(\mathbf{r}, \omega) - \Phi(\mathbf{r}, \omega_0))$. When $\Phi(\mathbf{r}, \omega)$ is Taylor-expanded to first order in frequency around ω_0 , i.e. $\Phi(\mathbf{r}, \omega) \simeq \Phi(\mathbf{r}, \omega_0) + (\omega - \omega_0) \partial_{\omega_0} \Phi(\mathbf{r}, \omega)$, the condition for constant envelope phase is found to be

$$\delta(\Phi_{\text{envelope}}) = 0; \quad (\text{S6})$$

therefore, we have

$$\begin{aligned} (\omega - \omega_0) \delta(t - \partial_{\omega_0} \Phi(\mathbf{r}, \omega)) &= (\omega - \omega_0) (\delta t - \\ \partial_{\omega_0} \nabla \Phi(\mathbf{r}, \omega) \cdot \delta \mathbf{r}) &= 0. \end{aligned} \quad (\text{S7})$$

Defining a unit vector for the position of the envelope, $\hat{\mathbf{r}} = \delta \mathbf{r} / \delta r_g$, then gives

$$\begin{aligned} v_g &= \frac{\delta r_g}{\delta t} \\ &= \frac{1}{\hat{\mathbf{r}} \cdot \nabla \partial_{\omega_0} \Phi(\mathbf{r}, \omega)} = \frac{1}{|\nabla \partial_{\omega_0} \Phi(\mathbf{r}, \omega)|}. \end{aligned} \quad (\text{S8})$$

The amplitude of a Laguerre-Gauss beam is given by

$$\begin{aligned} \text{LG}_{p,\ell}(\mathbf{r}, \varphi, z) &= \\ \frac{C_{p,\ell}}{w(z)} \left(\frac{r\sqrt{2}}{w(z)} \right)^{|\ell|} \exp\left(-\frac{r^2}{w^2(z)}\right) L_p^{|\ell|} \left(\frac{2r^2}{w^2(z)} \right) e^{i\Phi^{\text{LG}}}, \end{aligned} \quad (\text{S9})$$

where p is the radial index (positive integer), ℓ is the azimuthal index (integer), r and φ are the transverse cylindrical coordinates, $C_{p,\ell}$ are normalization constants, $w(z) = w_0(1 + (z/z_R(\omega))^2)^{1/2}$ is the beam radius, $L_p^{|\ell|}(x)$ are the generalized Laguerre polynomials, and Φ^{LG} contains all of the phase terms of the Laguerre-Gauss beam. The LG beam phase may be expressed explicitly as

$$\Phi_{p,\ell}^{\text{LG}}(\mathbf{r}, \varphi, z, \omega) = \frac{\omega}{c} z + \frac{\omega}{c} \frac{r^2}{2R(z, \omega)} + \ell \varphi - (2p + |\ell| + 1) \zeta, \quad (\text{S10})$$

where $\zeta := \arctan\left(\frac{z}{z_R(\omega)}\right)$ is the Gouy phase. The spatial dependence of the phase profile of the Laguerre-Gauss beam is the origin of the other-than-luminal propagation velocity. The Gouy phase is an obvious example of a non-trivial term in the phase of a beam that will give rise to sub- and super-luminal group velocities. By comparing the surfaces of constant phase of the Gaussian beam along with those associated with a spherical wave, one can observe how the phase of a Gaussian beam differs from that of a spherical wave. This difference suggests a discrepancy in the waves' group velocities as well. This concept is illustrated in Fig. S1, which shows the spatial dependence of the phase-fronts associated with Gaussian and spherical waves. This phase velocity leads to the conclusion that v_g is a function of r, z, ω, p and ℓ , i.e. $v_g(r, z; \omega; p, \ell)$.

Fig. S1. Phase distribution of Gaussian and spherical waves upon propagation in vacuum. **A** Comparison between spherical (dashed orange) and Gaussian (solid blue) wave phase-fronts having identical wavelengths. As the two beams propagate away from the focus, the phase fronts of the spherical and Gaussian waves begin to separate. This accrued phase discrepancy is associated with the Gouy phase [12]. **B** Illustration of the differing wavefront distributions of two Gaussian beams (dashed orange and solid blue, respectively) differing in wavelength by 2%. Insets show enlarged views of the phase-fronts near the beam waist (black colour box) and approximately at 10 wavelengths from the waist (gray colour box). The radial dependence of the difference between the phase-fronts associated with the two beams is apparent in both figures. This can be understood arise as a consequence of diffraction; the longer wavelength (orange) beam diverges more rapidly than the shorter wavelength (blue).

The radius of curvature $R(z, \omega)$ is obtained from the ABCD law for the case of a Gaussian beam incident on a thin lens at its waist, and subsequently focused to a new waist, and is given by [3]

$$R(z, \omega) = \frac{\left(\frac{z+d(\omega)}{z_{R,0}(\omega)}\right)^2 + \left(1 - \frac{z+d(\omega)}{f}\right)^2}{\left(\frac{z+d(\omega)}{z_{R,0}(\omega)}\right) - \frac{1}{f}\left(1 - \frac{z+d(\omega)}{f}\right)}, \quad (\text{S11})$$

where $d(\omega) = f \left(1 + (f/z_{R,0}(\omega))^2\right)^{-1}$ is the distance between the lens and the focus, f is the focal length of the lens, and $z_{R,0}(\omega)$ is the Rayleigh range of the beam prior to the lens. From the expression (S10) for the phase, one can obtain an explicit form for the group velocity of an LG beam by direct application of Eq. (S8),

$$v_g(\mathbf{r}, \omega) = \frac{1}{|\nabla \partial_\omega \Phi_{p,\ell}^{\text{LG}}(\mathbf{r}, \omega)|}. \quad (\text{S12})$$

This was the approach taken to produce the theoretical curves shown in Fig. 1 for LG modes having different indices p and ℓ . The group velocity associated with a pulse exhibits a dependence on $\mathbf{r}$. The radii at which velocity measurements were made correspond to the coordinates through which the classical rays associated with each beam will pass, according to the ray tracing picture. Measurements plotted in Fig. 3 naturally reflect the group velocity of LG modes at $r_{\text{max}} = w(z)\sqrt{|\ell|/2}$ for $p = 0$ modes having different values of ℓ .

We note also that two LG beams characterized by equal central wavelengths and r_{max} values, but different indices ℓ and, correspondingly, different beam waists, will propagate at different group velocities. Even if the classical ray trajectories associated with two LG modes are made to coincide at the focus by changing the beam waist, their Rayleigh ranges will differ, and scale inversely with ℓ . Therefore, it is impossible to compensate for this effect by simply changing the beam waist; it is a beam property rather than a property of the optics. It should also be noted that the results shown in Fig. 1 are wavelength and beam waist dependent.

Fig. S2. Time delay between LG mode pulses and a plane wave pulse as a function of position along the beam axis. **A** Theoretical arrival times of $\text{LG}_{p,0}$ pulses relative to a plane wave pulse travelling at c , as a function of position along the beam axis. Since the maximal intensity of $\text{LG}_{p,0}$ modes is found on-axis (at $r = 0$), these relative arrival times do not include any contribution from the phase-front radius of curvature. As a result, any deviations from exactly luminal propagation may be attributed entirely to changes in the p index itself. **B** Theoretical plot showing arrival times of $\text{LG}_{0,\ell}$ pulses as a function of propagation distance, relative to a plane wave pulse travelling at c . The time delays between LG pulses with nonzero ℓ indices and the plane wave pulse are monotonically-increasing functions of propagation distance for $\ell \neq 0$. This is due to the fact that LG beams with nonzero ℓ are characterized by subluminal group velocities throughout propagation. In each case, the plotted time delays are calculated at the radius of maximum intensity, $r_{\text{max}}(z)$, and therefore include

the contribution of the phase-front radius of curvature to the group velocity of the LG pulse.

Hypergeometric-Gaussian modes

A given Hypergeometric-Gaussian mode with well-defined ℓ can be regarded as an infinite superposition of $\text{LG}_{p,\ell}$ modes with fixed ℓ and different p indices. In a general, a HyGG beam generated from a pitch-fork hologram finds most of its power peaked in the $\text{LG}_{0,\ell}$ mode [4]. Perhaps the most important correction that one needs to include while dealing with HyGG modes is the radius of maximum intensity r_{max} . In the following, we carry out a calculation to obtain an expression for the r_{max} of a HyGG beam at the focus.

The HyGG modes (in dimensionless coordinates) can be expressed as a superposition of modified Bessel function of the first kind.

$$u_{-|m|,m}(\rho) \propto e^{\frac{1}{2}\left(\frac{-2i}{1+\zeta^2} + \frac{1}{i\zeta+\zeta^2}\right)\rho^2} \rho \left[I_{\frac{|m|-1}{2}}\left(\frac{\rho^2}{2\zeta(1+i\zeta)}\right) - I_{\frac{|m|+1}{2}}\left(\frac{\rho^2}{2\zeta(1+i\zeta)}\right) \right],$$

where $\rho = r/w_0$ and $\zeta = z/z_R$ are the dimensionless radial and longitudinal coordinates, respectively. We consider here only the radial dependence of the mode (for reasons that will be obvious as we proceed). The squared modulus of the modal amplitude is then calculated,

$$|u_{-|m|,m}(\rho)|^2 \propto e^{\left(\frac{\rho^2}{1+\zeta^2}\right)} \rho^2 \left| I_{\frac{|m|-1}{2}}\left(\frac{\rho^2}{2\zeta(1+i\zeta)}\right) - I_{\frac{|m|+1}{2}}\left(\frac{\rho^2}{2\zeta(1+i\zeta)}\right) \right|^2.$$

At this stage in the derivation, we assume that the term in the argument of the Bessel functions is very small. This condition is satisfied as the beam propagates away from the near field, at larger values of z . We can then approximate the modified Bessel function by,

$$I_\alpha(z) \approx \frac{1}{\Gamma(\alpha+1)} \left(\frac{z}{2}\right)^\alpha.$$

This approximation is valid in the limit where

$$0 < \left| \frac{\rho^2}{2\zeta(1+i\zeta)} \right| \ll \sqrt{\frac{|m|+1}{2}}.$$

The intensity of the beam can then be expressed in the following form,

$$|u_{-|m|,m}(\rho)|^2 \propto e^{\left(\frac{\rho^2}{1+\zeta^2}\right)} \rho^2 \frac{1}{\Gamma\left(\frac{|m|+1}{2}\right)} \left(\frac{\rho^2}{4\zeta\sqrt{1+\zeta^2}}\right)^{|m|-1} \left[1 - \frac{1}{|m|+1} \left(\frac{\rho^2}{1+\zeta^2}\right) + \left(\frac{1}{|m|+1}\right)^2 \frac{\rho^4}{4(\zeta^2+\zeta^4)} \right].$$

We proceed with an additional approximation, by neglecting the last term in the expression above. This is physically reasonable since z is assumed to be large in this case. Hence,

$$|u_{-|m|,m}(\rho)|^2 \propto e^{\left(\frac{\rho^2}{1+\zeta^2}\right)} \rho^2 \frac{1}{\Gamma\left(\frac{|m|+1}{2}\right)} \left(\frac{\rho^2}{4\zeta\sqrt{1+\zeta^2}}\right)^{|m|-1} \left[1 - \frac{1}{|m|+1} \left(\frac{\rho^2}{1+\zeta^2}\right) \right].$$

If we concern ourselves only with the radial dependencies,

$$|u_{-|m|,m}(\rho)|^2 \propto e^{\left(\frac{\rho^2}{1+\zeta^2}\right)} \rho^{2|m|} \left[1 - \frac{1}{|m|+1} \left(\frac{\rho^2}{1+\zeta^2}\right) \right].$$

We can now differentiate the expression in order to obtain the maximal value for ρ ,

$$\partial_\rho |u_{-|m|,m}(\rho_{\text{max}})|^2 = 0, \\ \rho_{\text{max}} = |m|^{1/4} (|m| + 1)^{1/4} \sqrt{1 + \zeta^2}.$$

Thus we obtain an expression slightly different from the case of LG beams, for which

$$r_{\max}^{(\text{HyGG})}(\ell, z) = w(z)(|\ell|(|\ell| + 1))^{1/4}.$$

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