Photopolymerization with high-order Bessel light beams

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We study photopolymerization with high-order Bessel light beams with phase singularities on-axis. Self-trapping and self-focusing of propagation-invariant light beams in a photopolymer allow the fabrication of extended helical microfibers with a length scale of a centimeter, which is more than an order of magnitude larger than the propagation distance of the Bessel light beams. We show the evolution of microfibers rotating at a rate proportional to the incident optical power, while the periodicity of the helical structures remains constant, irrespective of the laser power. This suggests that optical momentum transfer plays a predominant role in the growth and rotation of such fiber structures.

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Photopolymerization is the light-induced process of reacting monomer molecules to form polymer chains of a three-dimensional network within light-curing resins. One of the important classes of photopolymerization is the creation of self-written waveguides (SWWs) [1]. When exposed with low average optical power, self-trapping and self-focusing of light beams or optical solitons can occur along the beam propagation axis, leading to the formation of microfiber structures [2]. SWWs can have wider implications for the design of integrated optical circuits and devices in optical sensing and communications [3,4] as well as for the rapid fabrication of high aspect ratio microstructures without beam scanning [5].

Recently we demonstrated photopolymerization with Laguerre–Gaussian (LG) beams carrying orbital angular momentum (OAM) in the self-focusing regime [6,7]. Due to the presence of a phase singularity on the beam axis, LG beams exhibit an annular intensity profile in the direction transverse to the beam propagation axis. Such beams have gained prominence due to a diverse range of applications, e.g., in optical manipulation [8–10] and materials processing [11–13]. When launched into a self-focusing photopolymer, photopolymerization creates an optical vortex-soliton with a spiral trajectory [14], which results in an associated helical microfiber with a chirality determined by the sign of . Within the paraxial limit, our theoretical model, based on the nonlinear Schrödinger wave equation for the slowly varying electric field due to the permanent refractive index change [2], captures the main features of the experiment. These fabricated helical microfibers have the potential to provide new devices for mode generation, sorting, and optical communications, all based on OAM modes [15–17]. However, a major drawback to these applications is the limited length of these fibers (<200 μm) when created using LG modes [6]. Furthermore, the detailed mechanism of the growth of such fiber structures, specifically the role of optical forces acting on solidified polymers, remains unclear, and no quantitative conclusions have been made.

In this Letter, we investigate photopolymerization in response to high-order Bessel beams (BBs) possessing OAM at the visible wavelength of 532 nm. BBs are propagation-invariant beams, often termed “diffraction free,” which can persist for distances much longer than the Rayleigh limit. To date, zeroth-order BBs have demonstrated a successful fabrication of microfiber structures without a twist [18–23]. Here we launch the first-order BBs with a propagation range of >700 μm in a photopolymer. Self-trapping and self-focusing of the BBs lead to the formation of optical vortex-solitons and associated helical microfibers with a length scale of a centimeter, which is more than one order of magnitude larger than the propagation distance of the BBs. The rotation of microfibers is observed at a rate (up to 30 Hz) proportional to the incident optical power, while the periodicity of the helical structures, i.e., helical density, remains constant, irrespective of the laser power. This suggests that optical momentum transfer plays a predominant role in the growth and rotation of such fiber structures in a photopolymer. High-order BBs offer significant advantages for the fabrication...
of helical microfiber structures over extended ranges of $\geq 1$ cm when compared to LG light beams. Helical fibers of this length would be a significant step forward towards real applications in optical communications with OAM modes [24].

To obtain a high-order BB, we illuminate a conically shaped optical element termed an axicon with a LG light beam of order $\ell$, which transforms this beam into an approximation to a high-order BB of order $\ell$ [25]. Figure 1 shows a schematic diagram of the experimental setup. A linearly polarized continuous-wave laser with a wavelength of 532 nm is converted to a LG beam of order $\ell = 1$ by a spiral phase plate (SPP) followed by the axicon (apex angle 175°). The generated high-order BB is relayed by a set of lenses (L) to a microscope objective (MO) inside a sample cell containing a resin (NOA63) to initiate photopolymerization, which is monitored by a microscope objective (MO) inside a sample cell, the incident beam is focused inside the resin away from the self-trapping of the propagation invariant distance of the beam. Thus, optical element termed an axicon with a LG light beam of order $\ell$ (Fig. 1) can be avoided by adjusting the optical power (Fig. 1) so that only the central annulus performs photopolymerization. When the resin is exposed with a low average optical power ($0.7 \leq P \leq 0.9$ W), photopolymerization induces a positive refractive index change of the resin at a lag time or a critical exposure time $\tau_c$ (between the application of the laser field and the appearance of the axicon) which typically occurs within a period of 5 s $\leq \tau_c \leq 25$ s (depending on the optical power) from $n_{\text{unc}} = 1.52$ up to $n_{\text{cut}} = 1.56$ (when completely polymerized). Multi-ringed polymerization due to the optical power from $n_{\text{unc}} = 1.52$ up to $n_{\text{cut}} = 1.56$ (when completely polymerized) from $n_{\text{unc}} = 1.52$ up to $n_{\text{cut}} = 1.56$ (when completely polymerized) from $n_{\text{unc}} = 1.52$ up to $n_{\text{cut}} = 1.56$ (when completely polymerized).

Figure 2 shows combined line spectra, where each spectrum (or row of the image) represents scattered light intensities along the beam (z) axis, measured at a time between 11.7 s and 13.7 s (corresponding to the vertical axis) with a resolution of 2 ms. This time-lapse image of the fiber indicates that photopolymerization starts at around $t = 12$ s (approx. $t_1$), and the fiber extends in both forward and backward directions along the beam axis due to the presence of the back-scattered light guided through the fiber itself from both fiber ends [18]. Figure 3(d) shows the variation of the total scattered light intensity (blue curve) during the light exposure for 15 s, which is obtained by the sum of each row’s elements in Fig. 3(c). The rapid increase of helical microfiber structures over extended ranges of $\geq 1$ cm when compared to LG light beams. Helical fibers of this length would be a significant step forward towards real applications in optical communications with OAM modes [24].

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in the scattering intensity indicates the critical exposure time \( \tau_c \approx 12 \text{ s} \). The transmitted light intensity [orange curve in Fig. 3(d)] measured by the single pixel intensity in the phase singularity of the incident BB [Fig. 3(e)] illustrates the rotation of the microfiber [Fig. 3(f)], as previously reported in Ref. [6]. The fast intensity modulation coincides with the progression of photopolymerization, where a mean rate of rotation \( \Omega \approx 20 \text{ Hz} \) is measured during a period of \( 11.8 \leq t \leq 13.4 \text{ s} \). See also Visualization 1. Figure 3(g) shows the critical exposure time \( \tau_c \), required for photopolymerization and (h) mean rotation rate \( \Omega \) and helical density of microfibers for different incident optical powers. Experimental data of \( \tau_c \) and \( \Omega \) with error bars of \( \pm 2 \sigma \) are fitted with linear regression lines. (i) Time-lapse images of the transmitted light intensity pattern of LG \((P = 0.5 \text{ W})\) and BB \((P = 0.8 \text{ W})\), where \( \tau_c \approx 12 \text{ s} \). The scale bar indicates 5 \( \mu \text{m} \) and applies to all panels.

Figure 3(i) compares the transmitted light intensity pattern of LG \((P = 0.5 \text{ W})\) and BB \((P = 0.8 \text{ W})\), where \( \tau_c \approx 12 \text{ s} \). The scale bar indicates 5 \( \mu \text{m} \) and applies to all panels.

Microfibers with a length in the range of 0.5 cm–1 cm with the microscope objective (NA = 0.42) used. Figure 4(a) shows a typical example of fabricated microfibers with a length of 5.8 mm and a mean diameter of 6 \( \mu \text{m} \) when irradiated with the incident optical power of 0.85 W for the duration of 15 s. The helical structure of the fiber can be identified in the cross-polarized image [Fig. 4(b)], where we find 39 dark segments representing the helical features. This yields the helical density of 6.7/\( \mu \text{m} \), which is within the error of \( \pm 2 \sigma \) at 0.85 W in Fig. 3(h). Figures 4(c) and 4(d) are expanded views of the dashed enclosed areas in Figs. 4(a) and 4(b), respectively, showing the details of the microfiber. Here we find that the pitch of the helical structure is not evenly spaced across the fiber. The original structure of the fiber may have been altered because of the vigorous acetone treatment after photopolymerization. To obviate this in future, one could for instance perform photopolymerization in a microfluidic channel, where the resin can be washed away by a steady flow of acetone to minimize deformation of the polymerized fiber. We also note that linear polarization breaks the cylindrical symmetry of the BB, which in turn affects the fabricated microfiber structures. The use of circular polarization may be preferable for improving the quality of the polymerized fiber. Nevertheless, the density of the helical features is measured (>10 samples for reliable statistics) for each collected microfiber polymerized for each optical power \((0.7–0.9 \text{ W})\). Figure 3(h) compares the helical density (purple squares) with the mean rotation rate \( \Omega \) (orange triangles) at different optical powers, where the density remains unchanged, while \( \Omega \) is proportional to the optical power. We note that the use of high-order BBs allows to imprint many helical features (typically \( \geq 50 \)) on a long extended fiber structure (\( \geq 1 \text{ cm} \)), thus yielding reliable measurements of the helical density compared with LG beam polymerization exhibiting two to three features on a short microfiber (<200 \( \mu \text{m} \)).
Recently, in addition to our theoretical analysis [6], Nagura *et al.* have proposed a plausible model for the formation of helical fibers in photopolymerization with LG beams [26]. This model assumes that nanometric coarse-grains of polymer are first generated at the focal plane of the beam, which are subsequently launched into the vortex light field, where the Rayleigh scattering theory is applied to calculate optical forces and torques on these particles and simulate their motion. Considering submicrometer Rayleigh particles launched in a beam with the complex field amplitude of \( E(r, \phi, z) = E_0(r, z) \exp(\imath \phi) \), both the \( z \) component of the gradient and scattering forces \( F_{\text{grad}, z} \) and \( F_{\text{cat}, z} \) and the azimuthal force \( F_{\text{cat}, \phi} \) scale with the field amplitude \( E_0^2 \) [26]. Assuming \( F_{\text{cat}, z} / F_{\text{grad}, z} > 1 \) so that the particles are driven by the beam along its propagation (positive \( z \)) direction and the azimuthal force as

\[
L_{p} = 2\pi r \frac{F_{\text{cat}, z} + F_{\text{grad}, z}}{F_{\text{cat}, \phi}}.
\]

This implies that the period of a helical trajectory is independent of laser power. The coarse-grained model supports Fig. 3(h) showing that the incident power of the laser has no effect on the periodicity of the helical structures, suggesting that optical forces acting on the solidified polymers are responsible for the growth and rotation of helical microfibers in BB polymerization. Although further theoretical and experimental work is required to validate the applicability of this approach, the Rayleigh scattering model may offer an alternative route to simulate the photopolymerization process, complementing the paraxial soliton numerical study we explored in previous work [6].

In conclusion, we have demonstrated photopolymerization with first-order Bessel light beams possessing OAM with phase singularities on-axis. Self-trapping and self-focusing of the propagation invariant light beams lead to the formation of optical vortex-solitons and associated helical microfibers with a length scale of a centimeter, which is more than an order of magnitude larger than the propagation distance of the BBs. The OAM of the light imprints spiraling features across microfibers, where the rotation rate (mean rate of 20 Hz) increases with the incident optical power, while the periodicity of the helical features or the helical density remains unchanged, irrespective of laser power. This suggests that optical momentum transfer plays a predominant role in the growth and rotation of such fiber structures in a photopolymer.

Additional dataset supporting this publication can be accessed at [27].

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