Theoretical model

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January 2, 2018

1 Theoretical model

We employ the same model and notation for propagation of UV light of frequency ω in a curing resin as described by Kewitsch and Yariv in Ref. [1]. Then the paraxial wave equation for the slowly-varying electric field envelope $E(\mathbf{r},t)$ propagating dominantly along the z-axis is

$$\frac{\partial E}{\partial z} = \frac{1}{2k_b}\nabla_T^2 E + ik_0 \Delta n'(\mathbf{r},t)E - \frac{\alpha}{2}E, \tag{1}$$

where $k_b = n_b k_0$, n_b being the background refractive-index, $k_0 = \omega/c$, ∇_T^2 is the transverse Laplacian describing beam diffraction, and the electric field is scaled such that $|E|^2$ is an intensity. Furthermore, $\Delta n'(\mathbf{r},t)$ is the real part of the index response due to the UV light-induced photopolymerization

$$\Delta n'(\mathbf{r},t) = \Delta n_0' \left(1 - \exp\left[-\frac{1}{U_0}\int_0^{t-\tau} |E(\mathbf{r},t')|^2 dt' \right] \right), \tag{2}$$

where τ is the monomer radical lifetime which dictates the time delay between the application of the laser field and the appearance of the optically-induced refractive-index change, and U_0 is the critical exposure needed for photopolymerization. In contrast to Ref. [1] we do not include the imaginary part of the index response but rather simply include single-photon absorption via the coefficient α (see below).

For the NOA63 sample considered and a UV wavelength of 405 nm we set $n_b = 1.52$, $\Delta n_0' = 0.04$, and the lag time was estimated from measurements as $\tau = 0.1$ s. It is known that in the wavelength range $350 - 380$ nm the critical exposure is around $U_0 = 4.5$ J/cm^2. However, the single-photon absorption is much smaller at 405 nm and we estimate based on absorption measurements that $U_0 = 500$ J/cm^2 under the conditions of our experiment.

Finally, due to the smallness on the single-photon absorption we neglect it ($\alpha = 0$) over the 200 μm propagation lengths considered.

We have numerically solved Eqs. (1) and (2) using the Beam Propagation Method (BPM) for a variety of initial conditions corresponding to fields carrying OAM and characterized by their topological charge ℓ. Inspection of the inset panels in Fig. 2 reveals that the incident fields utilized in the experiment may be characterized as Laguerre-Gaussian (LG) beams [2] with topological charge ℓ, and associated radial mode index $p = (\ell + 1)$, so for $\ell \neq 0$ the corresponding intensity profiles are composed of concentric rings. In the numerics we have modeled the input beams as focused LG beams based on a Gaussian spot size $w_0 = 3$ μm, which in the range $\ell = 1 - 4$ gives radii of the ring of peak intensity in the range $1 - 2$ μm. In addition, the incident input power of 0.3 mW in the experiment translates to 0.1 mW at the sample surface and yields peak input intensities around $I_p = 1000 - 2000$ W/cm^2. Therefore, for the exposure time of around 1 s for the experiment an incident fluence of 1000 J/cm^2 exceeds the critical exposure $U_0 = 500$ J/cm^2, and the simulations are expected to display the photopolymerization process for the conditions of the experiment.

The results of the simulations are displayed as isometric plots of the induced real index change $\Delta n'(\mathbf{r}, t)$ evaluated at the exposure time $t = 0.9$ s, as this will reflect the formation of any induced waveguides during the photopolymerization process. Figure 7(a) is for $\ell = 0$ and shows the self-focusing and self-trapping previously reported in Ref. [1], with the formation of a single induced fiber with a high degree of cylindrical symmetry around the z-axis throughout the propagation. In contrast, Fig. 7(b) is for $\ell = 1$ and around $z = 100$ μm we see that the induced fiber breaks to one side and therefore clearly breaks the cylindrical symmetry around the z-axis. This behavior is similar to what is seen at the bottom of panel (b) in Fig. 3 from the corresponding experiment. In this context we remark in all of the simulations a 1% amplitude noise was added to the incident LG beams to mimic unavoidable imperfections in the input beams and seed any potential symmetry breaking present in the system. Without this added noise all of the simulations remained cylindrical symmetry, and the noise is what allowed for the breaking of the fiber to one side in Fig. 7(b). Finally, Figs. 7(c-e) show the main results of the simulations for $\ell = 2 - 4$, and clearly support the dominant features seen in the experiment: The initial vortex beams generate ℓ fibers that spiral around each other. For the case of $\ell = -4$, Fig. 7(f) illustrates that reversing the sign of the topological charge reverses the direction of the spiraling, compare with Fig. 7(e) for $\ell = 4$.

These simulations demonstrate that the main features of the experiment

are captured by the model of Kewitsch and Yariv [1], and it remains to provide a physical context for the observations. Previous studies of propagation in nonlinear Kerr media or saturable nonlinear media have shown that initial vortex beams with ring intensity profiles can indeed breakup into solitons via transverse Modulational Instabilities (MIs), and the solitons fly off tangential to the ring as demanded by conservation of OAM [3, 4, 5], akin to free particles. Such a transverse MI, seeded by the added noise, underlies the breakdown of the initial cylindrical symmetry seen in Figs. 7(c-e), where each initial vortex of topological charge $\ell = 2, 3, 4$ breaks into ℓ fibers: Changing the noise strength changes when the symmetry breaking sets in along the z-axis but the qualitative picture remains the same. However, in our case the fibers so formed are seen to spiral around each other as opposed to flying off tangential to the ring. The distinction is that the nonlinearity due to photopolymerization has a delay time $\tau = 0.1$ s which is much slower than the time scale on which the relative phase between the generated solitons evolves. This time delay renders the bright solitons created in the photopolymerization process mutually incoherent, and it is known that this leads to an attractive force between the solitons [6]. So the solitons and associated fibers generated in our photopolymerization experiment and simulations exhibit spiral trajectories due to the combined actions of the initial OAM supplied by the initial vortex, which on its own would lead to tangential motion, plus the attractive interactions between the solitons due to the mutual incoherence of the solitons, which pulls the solitons and fibers towards each other.

References

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