Control of the Refractive Index in Photopolymerizable Materials for (2 + 1)D Solitary Wave Guide Formation

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We report an experimental and theoretical study on the optimization of (2 + 1)D self-written waveguide formation inside a photopolymerizable material. The accurate control of the refractive index value inside the bulk of the material during the polymerization process gives us the opportunity to define a virtual core and a virtual cladding for the system. The $V$ value which characterizes the guidance properties of a fiber can be applied to this propagation. The control of the $V$ value allows us to propagate single mode or multimode waveguides on a few centimeters. Numerical simulations of these waveguides based on a paraxial model including both photopolymerization and Kerr effect give very good agreement with our experimental results.

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In photosensitive materials such as germano-silicate glasses or photopolymerizable resins, an exposure to light of appropriate wavelength produces a permanent increase of the refractive index which is a function of the absorbed optical energy. When these materials are excited with a suitable laser beam, the increase of the refractive index can compensate the natural diffraction, and the beam is thus self-trapped along the propagation axis. Under particular conditions this phenomenon can conduce to the formation of a permanent waveguide as reaction proceeds. This phenomenon is often referred to as light induced self-written (LISW) waveguides. It was described theoretically in 1995 by Monro et al. [1], and the first experimental demonstration came a few months later in a work reported by Kewitsch et al. [2,3].

Despite their potential applications in the field of integrated optics, LISW waveguides have not met practical applications yet. Indeed, the self-written process can be chaotic [4] and the inscription of uniform waveguides over lengths of a few centimeters is very sensitive to the experimental parameters. Recently, Kagami et al. [5] have proposed a refined technique based on a judicious blend of two kinds of photosensitive materials to create the core and the cladding of the guide. Because of this technique they obtained 20 mm long LISW waveguides with a propagation loss less than 1 dB/cm.

In this Letter, we report a detailed study on light induced self-written waveguides. Our work, which includes both experimental and theoretical aspects, has given to new elements a better understanding, leading to an optimization of the inscription of LISW waveguides. In the experimental section we discuss the influence of various physical parameters (such as light intensity, evolution of the refractive index of the medium upon irradiation) on the inscription of LISW waveguides. Drawing an analogy with the usual waveguides we have defined a $V$ value for the guides written in the photosensitive resin, and we have shown that $V$ is a convenient parameter to predict which type of propagation will occur. Then, a model based on the paraxial wave equation [1] has been used to describe the propagation of light in a photopolymerizable medium. Our experimental results led us to include new refinements in the model we developed, compared to previous studies [3,6,7,9,10]. In particular, we have demonstrated that the saturation of the photopolymerization and also an instantaneous focusing Kerr effect should be taken into account to obtain relevant numerical simulations.

For our experiments we worked on layers of resin 250 μm thick. These layers were formed by enclosing the photopolymerizable formulation between two glass slides separated by calibrated spacers. The tip of a single mode fiber was then introduced into the photopolymerizable layers and the 514 nm line of an argon ion laser was used to irradiate the sample through the fiber. The propagation of the light inside photosensible materials leads, under certain conditions, to the formation of spatial sol-

FIG. 1. Refraction index variation of the core (curve) and the cladding (curve).
itons [4]. Indeed, in this previous work, this propagation cannot be controlled because of the lack of knowledge of the behavior of the refractive index. Here, the formation of photopolymerized guides has been treated as optical fibers made by three parts: the core, the cladding, and the jacket, which has no optical utility. Our experimental work consisted of treating the medium with a linear cladding and a nonlinear core, which is the symmetrical theoretical problem investigated by Micallef and co-workers [11]. It is well known that, to carry out the guiding of the light in a fiber, the value of the refractive index of the core has to be higher than the cladding one. We have applied the same approach to our samples. This was possible using the results of a previous work allowing the measurement of the temporal dependence of the refractive index inside photopolymerizable materials [12] upon illumination. Using this method, it is possible to manage the difference of the refractive index values between the virtual core and cladding of the guide. First, the control of the value of the refractive index of the virtual cladding is obtained by a prepolymerization of the whole sample by a 50 W halogen lamp. The lower curve in Fig. 1 shows the evolution with time of the cladding refractive index upon this illumination. Then, after a specified prepolymerization which increases the refractive index of the cladding to a given value, the argon laser beam is introduced inside the sample using a single mode fiber to create the virtual core of the guide. Thus, the value of the refractive index of the core starts from the cladding refractive index value to reach the upper final value (see upper curve in Fig. 1). At this point, the spatial shape of the guide depends on the duration of the prepolymerization, or equivalently on the value of the refractive index of the cladding. One can obtain a single mode, a multimode guide, or even a number of distinct guides. We have chosen to refer to the usual $V$ value defined as $V = \frac{2\pi a}{\lambda} \sqrt{n_{\text{core}}^2 - n_{\text{cladding}}^2}$, where $a$ is the fiber diameter (about 10 µm), $n_i$ the refractive index region $i$, and $\lambda$ the wavelength (here 514 nm). This $V$ factor has to be less than the value of 2.4 in order to ensure a single mode propagation.

When the prepolymerization duration is higher than 40 min we are able to create a single guide (Fig. 2 top). In that case, $\Delta n = n_{\text{core}} - n_{\text{cladding}}$ is very weak, around $10^{-4}$ and $V < 2.4$. When the prepolymerization duration time is around 40 min, then the value for the $V$ factor is about five and we obtain two guides as shown in Fig. 3. Finally, if there is no prepolymerization or if the prepolymerization time is less then 15 min, then the corresponding $V$ factor is greater than 20 and we create chaotic guides (see Fig. 4). Indeed, by controlling the cladding illumination time, we can manage the difference between the refractive index of the core and the cladding, and therefore we can control the guide formation inside the cell. We also experimentally demonstrate that the $V$ factor is the sole relevant parameter needed to create single or multimode guides by photopolymerization.

**FIG. 2.** Control of the solitonic guide inside the sample. (a) Guide obtained after 1 h of illumination. (b) Top: simulation of light propagation in the guide; bottom: index variation $\Delta n$ versus transverse $x$ and longitudinal $z$ space variables, at a given time $t_0$, for a Gaussian input $u_0 = a/\exp(x/0.3)^2$. Parameters: $\alpha = 0$, $\gamma = 0.5$, $\Delta n_z = 30$, $t_0 = 0.6$, $a = 10$ (dimensionless values).

**FIG. 3.** Splitting of the solitonic guide inside the sample. (a) Guide obtained after 45 min of illumination. (b) Top: simulation of light propagation in the guide; bottom: index variation $\Delta n$. Same parameter as Fig. 2(b), except $t_0 = 0.14$, $a = 25$ (dimensionless values).
Let us now study the dynamics of the waveguide formation, in order to explain the waveguide splitting which occurs when the input power is increased: The analysis starts from the paraxial wave equation approximation as proposed by Monro et al. [6, 7, 9, 10]. We define the propagation direction along the $z$ axis, and consider one transverse spatial coordinate $x$. The evolution of the electric field envelope $E$ is, in a first approximation, governed by the equation

$$ik_0n_0\frac{\partial E}{\partial z} + \frac{1}{2}\frac{\partial^2 E}{\partial x^2} + k_0n_0\Delta n E + i\frac{1}{2}k_0n_0\alpha E = 0,$$

(1)

where $\alpha$ is the linear absorption coefficient, $k_0$ the wave vector in vacuum, $n_0$ the refractive index of the nonpolymerized material, and $\Delta n$ the change of the refractive index due to photopolymerization. It evolves according to

$$\Delta n = n_0\int_{-\infty}^{t} |E|^2 dt,$$

(2)

where $t$ is some normalized time. Using the transform

$$z = 2z', \quad t = \frac{d^2}{2k_0n_0}t', \quad x = \frac{x'}{\sqrt{k_0n_0}}, \quad E = \frac{E'}{d'}, \quad \Delta n' = \frac{\Delta n'}{2k_0},$$

(3)

we reduce Eqs. (1) and (2) to the normalized form:

$$i\frac{\partial E}{\partial z} + \frac{1}{2}\frac{\partial^2 E}{\partial x^2} + \Delta n E + i\alpha E = 0,$$

(4)

$$\Delta n = \int_{-\infty}^{t} |E|^2 dt.$$

(5)

An essential remark concerns the homogeneity of Eq. (4). If we set $E = d'E, t = t'/d^2$, Eqs. (4) and (5) are not modified, whatever the number $d$ is. The fact of increasing the light intensity only speeds up the process. We observed experimentally that the shape of the waveguide depends on the input intensity. The above mentioned homogeneity property shows that it cannot be accounted for by this model as it is. At least a second reference value for the intensity is needed, therefore another nonlinear effect must be taken into account. The possible nonlinear effects in the nonphotopolymerized medium could be either a Kerr effect, focusing or defocusing, or a nonlinear absorption. Other nonlinear effects can be related to the photopolymerization process, as a photopolymerization energy threshold, or a saturation. We considered each of these nonlinear effects, and perform numerical simulations of the simultaneous beam propagation and waveguide formation. The results obtained when either the defocusing Kerr effect, the nonlinear absorption, or the photopolymerization energy threshold are taken into account, do not reproduce the experimental results, while a very good agreement between theory and experiment is obtained when an instantaneous focusing Kerr effect is considered. We can thus conclude that the latter is present and responsible for the qualitative modification of the waveguide shape when the input power is increased. Let us detail the corresponding model: Eq. (4) is replaced by

$$i\frac{\partial E}{\partial z} + \frac{1}{2}\frac{\partial^2 E}{\partial x^2} + \Delta n' E + i\alpha E + \gamma|E|^2 E = 0,$$

(6)

where $\gamma > 0$ is proportional to a nonlinear third order susceptibility. Numerical computations show that, at low intensity, the formation of the guide is enhanced, while the beam breaks up when the intensity is increased. However, the saturation of the photopolymerization must be taken into account. The refraction index, indeed, cannot exceed the value $n_0 + \Delta n'$, corresponding to the completely polymerized material, while the model yielded by Eqs. (5) and (6) produces values of the index variation $\Delta n$ larger than $\Delta n'$. The corresponding equations are derived as follows. We denote by $P$ the initial number of photopolymerizable molecules, and by $p$ the number of molecules already photopolymerized. $\Delta n$ is apriori proportional to $p$, and $\frac{\partial \Delta n}{\partial t}$ to $(P - p)$. We thus obtain

$$\frac{\partial \Delta n}{\partial t} = \left(1 - \frac{\Delta n}{\Delta n_c}\right)|E|^2,$$

(7)
with \( \Delta n_{p} = \frac{\Delta n}{p} \). We notice that the Kerr effect, and thus the nonlinear coefficient \( \gamma \), must be proportional to the quantity of nonpolymerized matter, i.e., to \( (P - p) \), leading to \( (1 - \frac{\Delta n}{\Delta n_{p}}) \).

Therefore and for the sake of theoretical coherency, we take it into account in the model, which leads to an effective saturation of the Kerr effect. Thus, Eq. (6) becomes

\[
\frac{i}{\partial z} \frac{\partial E}{\partial z} + \frac{\partial^{2} E}{\partial x^{2}} + \Delta n E + i \alpha E + \gamma \left( 1 - \frac{\Delta n}{\Delta n_{p}} \right) |E|^{2} = 0. \tag{8}
\]

The effective saturation of the Kerr effect expresses the fact that only the monomer is assumed to produce it. The absence of focusing the Kerr effect in the polymer is by no means proven; we do not include it in the model because it is not necessary to retrieve the experimental results. In one spatial dimension, the saturation of the Kerr effect does not modify strongly the behavior, while in two dimensions, it is known to be able to prevent collapse. The fact that experiments in bulk do not show the collapse could be explained by the absence of focusing the Kerr effect in the polymer, and is thus an argument in favor of this assumption. This must be confirmed by further studies.

The simulations presented below are obtained by using the model Eqs. (7) and (8). For this we notice that the refractive index variation \( \Delta n \) evolves very slowly with regard to the light propagation; thus time intervenes in Eq. (7) as a parameter only, through \( \Delta n \). The principle of the numerical resolution of Eqs. (7) and (8) follows: At a given time \( t \), the electric field \( E \) is given by the propagation Eq. (7), where \( t \) does not appear. This field then writes a guide by modifying \( \Delta n \). At a further time \( t + \delta t \), light propagates in the guide, which began to be written, and so on.

At low input intensity, a regular “solitonic” waveguide is formed [Fig. 2(b)]. The index variation \( \Delta n \) presents longitudinal oscillations. When the input power is increased, the light diffused by these inhomogeneities becomes strong enough to initiate the formation of two secondary waveguides. No light goes into the central guide any more, which stops its formation [Fig. 3(b)].

When the power is further increased, this phenomenon repeats, and other lateral guides are formed. The evition of light from the previous guides is much less efficient as before, which leads to the chaotic aspect of Fig. 4(b).

In conclusion, we have experimentally explored the different stages of guide propagation in bulk photopolymerizable media. Computer simulations based on a paraxial propagation equation also corroborated these studies. The growth of both single-channel and multichannel guides depends on the duration and the power of the input light. The permanent properties of the guide induced in such photopolymerizable mixtures open new possibilities to develop organic optical devices. By controlling the shape of the guides it might be possible to draw optical circuits at a microscopic scale. Moreover, by doping the guides with active nonlinear optical chromophores, various optical functions might be realized.

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