

LASER INDUCED SELF-ACTION PHENOMENA IN A PHOTOPOLYMERISABLE MEDIUM

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Abstract

The nonlinear propagation of a visible, continuous wave laser beam in a photopolymerizable organosiloxane was studied at intensities ranging across 10 orders of magnitude (3.2×10^{-5} to 12732 W/cm^2). The process was characterised in detail through spatial intensity profiles of the beam, temporal monitoring of its width and peak intensity combined with optical microscopy of the resulting self-induced structures.

These observations revealed a rich diversity of dynamic phenomena during nonlinear light propagation in different intensity regimes including (i) optical self-trapping (ii) *in situ* sequential excitation of high-order modes (corresponding to optical fiber modes) in self-written cylindrical waveguides, (iii) variations in modal composition during the transition of self-written waveguides from single to multimode guidance, (iv) generation of spatial diffraction rings that propagated over long distances ($>$ Rayleigh length), (v) transformation of the Gaussian beam into an unstable single ring, which collapsed into azimuthally symmetric filaments and (vi) complete beam filamentation.

Extensive and quantitative analyses of spatial beam profiles provided insight into the mechanisms underlying each of these phenomena, particularly the significance of the spatial profile (gradient) of refractive index changes induced in the medium. The experimental findings were consistent with results of numerical simulations of nonlinear light propagation in the corresponding intensity range that were implemented through the beam propagation method with the software BeamPROPTM. The results of these comprehensive series of experimental and theoretical studies provide a deep understanding of the dynamics of nonlinear light propagation in a photopolymerizable medium and are consistent with some predictions of earlier theoretical models.

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Chapter 1

Introduction

Optical beams that alter their own path as they travel through a medium lead to a variety of self-action phenomena. Conditions under which these phenomena occur are classified as nonlinear because light propagation is described by nonlinear differential equations. The best known example is the self-trapped beam or optical soliton, which suppresses its own diffraction (or divergence) by modifying the refractive index of the medium through which it propagates.

Originally, optical self-action effects have been related with high laser input intensities in glasses and atomic vapours or more generally Kerr media. Progressively, other media have been employed including photorefractives, liquid crystals and photopolymers that do not require high intensities to observe similar effects. The photoreponses of different media, however originate from different mechanisms and therefore new dynamics and effects of nonlinear propagation have been discovered.

Self-action effects are categorized as nonlinear because they can be described by nonlinear differential equations. In the case of most nonlinear optical materials (NLO) such as Kerr media and photorefractive crystals, they are called nonlinear because their photoinduced refractive index changes originate from high order (nonlinear) susceptibility terms. In the case of photopolymers, a nonlinear propagation of light originates due to refractive index changes induced by photochemical reactions, and such propagation can also be described by a nonlinear differential equation (*vide infra*).

In this Chapter we will introduce optical self-action phenomena with emphasis on those effects that modify the natural diffraction of beams. In the following Sections we will review optical nonlinearity in a variety of media and self-action effects that will be relevant to the studies in this thesis including: optical self-trapping and spatial solitons, spatial self-phase modulation and filamentation due to modulation instability. Finally, the research objectives and presentation of this thesis will be provided.

1.1 Optical nonlinearity in various media

The field of nonlinear optics has been studied for almost 50 years [1] and is concerned with the nonlinear response of materials interacting with electromagnetic fields. The beginnings of nonlinear optics go back to the invention of the laser in the

late fifties by Townes and Schwlow [2], since traditionally high intensities have been required to excite nonlinear responses in optical materials such as glasses [3], absorbing liquids (presenting thermal nonlinearity) [4], atomic vapours [5], liquid crystals [6] and semiconductors [7].

Nonlinear optical (NLO) materials have been divided in different classes based on the effects that they present, the two most widely known are Kerr and photorefractive effects. The photoresponse of these and most other NLO materials rely on the nonlinear dielectric susceptibility response upon light exposure. Photopolymers, have recently been investigated, which rely on photochemical reactions for changes in the refractive index to take place.

In the following Sections, we will review some of the mechanisms of materials that rely on high order susceptibilities for their nonlinear photoresponse. The photoresponse in photopolymers will be reviewed with emphasis on an organosiloxane medium, which is the medium being investigated in this work. Comparison between the photoresponses of various NLO media and photopolymers will be made.

1.1.1 Various nonlinear optical mechanisms

Most optical nonlinear materials rely on the motion of electrons as a response to an electric field (stationary or varying) for their nonlinearity. This motion leads to a bulk material polarization of the following form [1]

$$P = \epsilon_0 (\chi^{(1)} E + \chi^{(2)} E E + \chi^{(3)} |E|^2 E + \dots) \quad (1.1)$$

where ϵ_0 is the permittivity in vacuum, E is the electric field amplitude and $\chi^{(n)}$ is the susceptibility with $n=1, 2, 3, \dots$ order. The first term in Equation (1.1) corresponds to the linear response of the medium to the electric field associated with an optical beam, the second and third term correspond to a nonlinear response related to a weak asymmetric and symmetric anharmonicity with respect to the electron displacement, respectively.

Equation (1.1) sets the basis for the photoresponse of the principal media employed in studies of spatial self-action phenomena including Kerr, photorefractive nonlinearity and quadratic nonlinearity.

Kerr nonlinearity

The nonlinear polarization giving rise to the Kerr effect is $P_{NL} = \epsilon_0 \chi^{(3)} |E|^2 E$, the third term in Equation (1.1). The resulting refractive index associated with the nonlinear response can be written as $n = n_0 + n_2 |E|^2$ [8], where n_0 is the linear or background refractive index and n_2 is the Kerr nonlinear refractive index coefficient. The second term is the refractive index change which is dependent on intensity $\Delta n = n_2 |E|^2$ and which is responsible for the observed self-action phenomena. This particular form of the index change has played an important role in nonlinear optics

as it allows for analytical solutions of various problems, one example is in 1-D spatial solitons as will be discussed in Section 1.2. Since the larger the nonlinearity is, the slower the response (turn-off) time becomes [9], Kerr nonlinearity being the weakest one possesses a virtually instantaneous (10^{-15} s) and local nonlinearity.

Although all materials exhibit a non zero $\chi^{(3)}$, only few of them are used in nonlinear optics for having a measurable Kerr response. This corresponds to a linear dependence of the index change on intensity, at intensities useful for nonlinear effects. Two examples are the semiconductor AlGaAs and glass fused silica with $n_2 = 1.5 \times 10^{-13}$ cm²/W and $n_2 \approx 2.3 \times 10^{-16}$ cm²/W, respectively [9]. As the nonlinear coefficients are small, high powers (intensities) on the order of kilowatts (GW/cm²) [10] are required for refractive index changes to occur (usually on the order of $\Delta n = 10^{-5}$ [3]). This is achievable with ultra-short laser pulses on the order of femtoseconds. As refractive index changes can grow indefinitely with intensity, Kerr materials are nonsaturable which often results in material damage during self-action effects at extremely large intensities.

Photorefractive nonlinearity

The nonlinear polarization induced in photorefractive media is given by $P_{NL} = \epsilon_0 \chi^{(2)} E_1 E_2$ corresponding to the second term in Equation (1.1). This can result in the electro-optic effect (Pockels), if an externally applied DC electric field (for example, $E_2 = E_0$) produces a polarization at the optical frequency (that of E_1) and modifies the refractive index in the medium as [8]

$$\Delta n_0 = -\frac{1}{2} n_0^3 \tilde{r} \cdot E_0 \quad (1.2)$$

where $\tilde{r} \propto \chi^{(2)}$ is the third rank electro-optic tensor that reflects crystalline symmetry and n_0 is the unperturbed refractive index of the medium in the relevant direction. This effect is only possible in non-centrosymmetric media.

Photorefraction resulting from charge transport occurs in specifically-doped electro-optic materials, which are typically dielectric or semiconductor crystals (e.g. SBN (Strontium Barium Niobate), LiNbO₃, KNbO₃ (Potassium niobate), KLTN (Potassium lithium tantalate niobate), InP, CdZnTe) [9]. These crystals contain a considerable level ($\sim 10^{18}$ cm⁻³) of donor impurities with energies deep in the forbidden gap, and a smaller level ($\sim 10^{16}$ cm⁻³) of deep acceptors. Therefore, illuminated regions generate an out-of-equilibrium concentration of free electrons that diffuse from regions of high concentration to those with lower ones. After some time (dielectric relaxation time), charges are re-trapped by localized ions leading to a space-dependent charge distribution, and hence to a space-charge field, which modifies locally the refractive index through the Pockels effect. As the space charge field is not uniform in space, the refractive index also varies in space [11]. The most commonly used

nonlinearity in photorefractive media is the screening nonlinearity, which yields an intensity-dependent index change of the form [9]

$$\Delta n(I(r)) = \frac{\Delta n_0}{1 + (I(r)/I_b)} \quad (1.3)$$

where $I(r)$ is the intensity of the optical beam and I_b is the background intensity which could be either dark irradiance or a material parameter proportional to the conductivity of the crystal in the dark. Δn_0 is the refractive index change due to Pockels effect shown in Equation (1.2). It can be more specifically expressed as $\Delta n_0 = -\frac{1}{2}n_0^3 r_{eff} V/L$, where r_{eff} is the relevant component of the electro-optic tensor, V the voltage and L the distance between the electrodes. Δn_0 is intrinsically saturable because when the space-charge field reaches equilibrium, no further index changes occur. The response time of the medium corresponds to the dielectric relaxation time (or necessary time for charge separation), which is inversely proportional to intensity [12]. By varying the intensity, the response time can be changed from nanoseconds to minutes for high and low intensities, respectively [13]. Powers (intensities) required for self-action effects are of the order of microwatts (mW/cm^2) for low response times and kilowatts (MW/cm^2) for faster response times. Refractive index changes are of the order of 10^{-3} [12]. The applied voltage over distance V/L is of the order of few kV/cm for both low and high intensities [13]. Being the first saturable media employed in the study of spatial solitons, photorefractive crystals have been relevant in this area as new type of solitons have been discovered along with other novel effects (Section 1.2).

Liquid crystal nonlinearity

Liquid crystals possess an intermediate phase between a crystal and a fluid. Various phases exist but the most used one in nonlinear optics is the nematic phase where the liquid crystal molecules are composed of elongated rod-like molecules. They exhibit an average orientational order and present an overall alignment towards a direction, called the director field ($\vec{n}$). Under these conditions, nematic liquid crystals are characterized by two indices of refraction: parallel ($n_{\parallel}$) and perpendicular ($n_{\perp}$) to their molecular axis, making them an optical uniaxial medium [14].

The nonlinearity originates from a reorientation of the molecules in the presence of an electric field (oscillatory or stationary), which can induce dipoles and therefore a torque between the nematic liquid crystals and the field itself. With a strong enough field, the director distribution can be changed, reorienting the molecules toward the electric field vector. The associated electric interaction energy density can be expressed as [15]

$$f_{el} = \frac{\epsilon_0 \Delta \epsilon}{2} \left\langle \left(\vec{n} \cdot \vec{E} \right)^2 \right\rangle \quad (1.4)$$

where $\Delta \epsilon = n_{\parallel}^2 - n_{\perp}^2$ is the birefringence and $\vec{E}$, the electric field vector. The minimization of f_{el} through reorientation corresponds to an index increase as the torque

tends to reduce the angle between the molecular axis and the field vector and a positive uniaxial is obtained ($n_{\parallel}^2 > n_{\perp}^2$) [15]. In the case when the field vector and the director ($\vec{n}$) are perpendicular to each other, the reorientation is subject to an intensity threshold, known as the Freedericks transition intensity.

The photoinduced Δn in nematic liquid crystals as seen in Equation (1.4) is intensity dependent, is saturable and has a strong polarization dependence. Δn is substantially independent of wavelength in the whole transparency range, from visible to mid-infrared regions [15]. The response time of nematic liquid crystals is slow, on the order of 10 to 100 milliseconds [16] as re-orientation of molecules in a fluid occurs through elastic restoring forces. The refractive index change can be in the order of 10^{-1} [16] and the typical power (intensity) scale required for self-action effects is in milliwatts (W/cm^2) [15].

Quadratic nonlinearity

All the previous nonlinearities present intensity dependent refractive index changes. However, self-action phenomena and more specifically self-trapping has also been observed in quadratic materials where no refractive index change or modification of the material is involved. Instead, self-action effects are enabled through strong interaction and energy exchange between two or more beams relying only on second order nonlinearities $\chi^{(2)}$ [8]. Therefore, this nonlinearity can only occur in non-centrosymmetric media in which phase matching is possible. From Equation (1.1) the induced nonlinear polarization in this type of materials is $P_{NL} = \epsilon_0 \chi^{(2)} E_1 E_2$, where E_1 and E_2 can possess similar or different frequency generating a third one, through parametric mixing processes. Self-action effects have been commonly observed during second harmonic generation [17] where one or two input fundamental fields (at the frequency ω) mix generating a second harmonic (2ω).

The quadratic nonlinearity is saturable, which can be understood by the conservation of the total electromagnetic energy, where if one beam increases in power, it can only do it at the expense of the other beam. Some quadratic media include KTP (potassium titanyl phosphate), LiIO_3 [18], LiNbO_3 and LiTaO_3 [9].

1.1.2 Photo-induced refractive index changes in photopolymers

Photopolymers in contrast to previous materials rely on photochemical reactions, specifically on free-radical polymerization for their refractive index change. A monomer is sensitized with a photoinitiator, which absorbs light, to produce reactive free radicals. A polymerization chain reaction is then started by reaction of the photoinitiated free radicals with the monomer functional groups [19]. The bonds formed during this process lead to a cumulative increase in local density and a corresponding increase in refractive index. Changes in refractive index are intensity dependent and

are determined by the rate equations that describe free-radical photopolymerization: rate of initiation and rate of propagation. The rate of initiation (r_i) is given by [19]

$$r_i = \Phi_i I_0 [1 - \exp(-\epsilon l [PI])] \quad (1.5)$$

where I_0 is the incident light intensity, l the sample thickness, ϵ and PI the absorptivity and concentration of the photoinitiator, respectively and Φ_i the quantum yield of initiation, which corresponds to the number of initiating species produced per photon absorbed. The rate of propagation (R_p) is also intensity dependent and can be expressed as [19]

$$R_p = \frac{k_p}{k_t^{0.5}} \{ \Phi_i I_0 [1 - \exp(-\epsilon l [PI])] \}^{0.5} M \quad (1.6)$$

where k_p and k_t are the rate constants of propagation and termination, respectively and $[M]$ is the monomer concentration.

The refractive index change associated with free radical polymerization at a particular location and time $\Delta n(x, y, z, t)$ in photopolymers has been found to be empirically related to the intensity of the optical field by Kewitsch and coworkers [20] by the following expression

$$\Delta n(x, y, z, t) = \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(t)|^2 dt \right] \right\} \quad (1.7)$$

where Δn_s is the maximum refractive index change, U_0 is the critical exposure required to initiate polymerization, τ is the monomer radical lifetime and $E(t)$ is the amplitude of the electric field. A plot of $\Delta n(x, y, z, t)$ with respect to radiant exposure as seen in Figure 1.1, shows the saturable nature of refractive index changes as monomers are depleted and the reaction reaches completion. Response time of these reactions range from milliseconds to seconds as polymerization proceeds and changes in refractive index changes can be as high as 0.15 [20]. Sensitization at a range of UV and visible wavelengths is achieved by choosing the appropriate photoinitiator. Intensities required for self-action effects in photopolymers can be as low as tens of $\mu\text{W}/\text{cm}^2$ [21]. Some of the monomers that have been employed for self-action phenomena include liquid diacrylate monomer [20], pentaerythritol triacrylate [22] and fluorinated epoxy monomer and acrylic acid and urethane-acrylate oligomer [23].

1.1.3 Organosiloxane structure

The material employed in our studies consists of an organic-inorganic composite which minimises diffusion of free-radicals commonly observed in pure monomer systems [24] that prevent localization of the induced refractive index change. Figure 1.2 shows the preparation and photopolymerization of the organosiloxane composites where polymerisable methacrylate groups are covalently bound to siloxane oligomers. As photopolymerization is initiated, methacrylate radicals remain attached to the

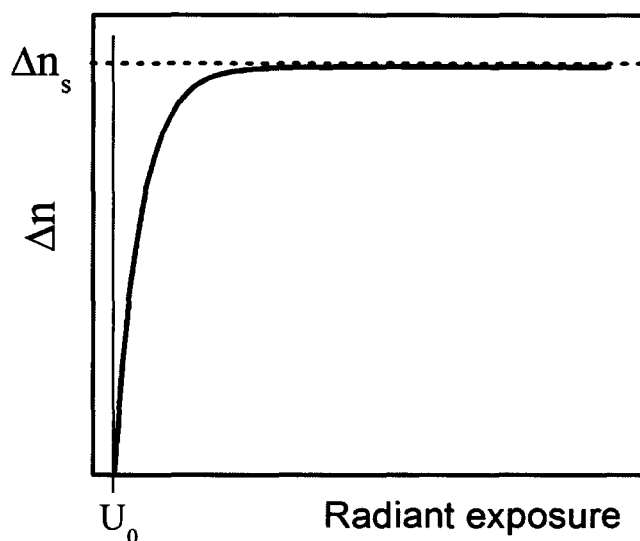


Figure 1.1: Plot of refractive index change Δn as a function of radiant exposure in photopolymers using Equation (1.7).

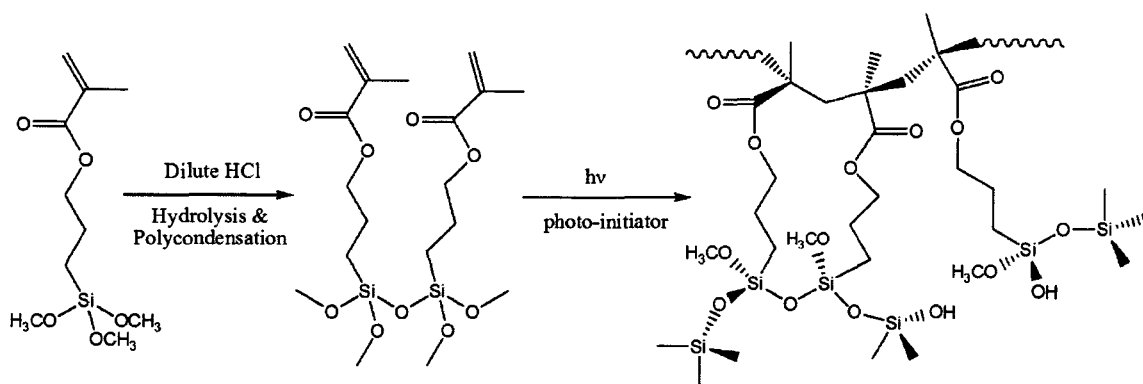


Figure 1.2: Scheme illustrating the preparation and photopolymerization of methacrylate-siloxane composites: the siloxane network is formed through hydrolysis and polycondensation, addition of a photoinitiator and light exposure starts the free-radical photopolymerization reaction. Illustration taken from [26]

siloxane network, this minimises diffusion of free-radicals causing the resulting refractive index strongly localized in space with a spatial resolution of ~ 150 nm [25].

1.1.4 Comparison of optical nonlinear mechanisms

The various mechanisms underlying the nonlinear photoresponse, specifically the refractive index change of materials determine the dynamics of self-action phenomena. It is important to point out similarities and distinguish differences between the various nonlinear materials and photopolymers to be able to compare self-action effects and understand the new phenomena that can arise.

A common feature of Kerr, photorefractive, liquid crystal photoresponse and photopolymers is their intensity-dependent refractive index change, even though the specific dependences are different as discussed in the previous Sections. One can divide the nonlinear photoresponse in saturable as in the case of photorefractive, liquid crystal nonlinearities and photopolymers and non-saturable like the Kerr nonlinearity. In saturable nonlinearities a maximum value of Δn is reached and in non-saturable media refractive index changes indefinitely with intensity, only hampered by damage of the material. This particular classification will become important when discussing self-trapped beams in Section 1.2.2 as self-trapping of circular beams (self-trapping in two directions) is only supported in saturable media.

One fundamental difference between most nonlinear optical media and photopolymers lies in the origin of the nonlinearity. In most media, refractive index changes arise from high order (nonlinear) susceptibilities that originate from the electron motion under the influence of an electrical field associated with an optical beam. In photopolymers, the nonlinear photoresponse originates in photochemical reactions that polymerize the material, densifying and changing its refractive index.

Another important difference derived from the origin of the nonlinear photoresponse, is that refractive index changes in photopolymers are permanent. Once the medium is polymerized, its structure permanently changes, and therefore optical structures can be formed. In other nonlinear media, index changes decay after the optical source is removed. This decay is dependent on response time, which is very fast for Kerr media (10^{-15} s), varies in photorefractives from nanoseconds to minutes depending on the dielectric relaxation time, and in liquid crystals is slow in the order of milliseconds due to the reorientation of the molecules. In photopolymers, the time for refractive index changes to occur depends on the monomer radical lifetime and the rate of the photopolymer reactions and it increases from milliseconds to minutes as the reaction proceeds. This slow reaction allows for the observation of incoherent self-trapping [27].

An advantage of photopolymers compared to other nonlinear media presenting intensity-dependent index changes, is that intensities required for self-action effects are much lower, of the order of $\mu\text{W}/\text{cm}^2$. In contrast, photorefractives require in-

tensities of at least three orders of magnitude higher and liquid crystals and Kerr media, six and fifteen orders of magnitude higher, respectively. Induced refractive index changes in photopolymers are at least two orders of magnitude larger than in Kerr and photorefractive media and comparable to those in liquid crystals.

1.2 Optical self-trapping and spatial solitons

The phenomenon of self-trapping of waves has been observed in many systems in nature including charge density waves in plasmas [28], sound waves [29] and electromagnetic waves [30]. The first reported observation of self-trapping of waves was in 1834 when John S. Russell observed that a smooth and well defined heap of water propagated through a canal without changing its form or decreasing its speed [31], he called it a *solitary elevation*. Theoretical studies on solitary waves in hydrodynamics were proposed later [32], followed by experimental and theoretical studies in plasmas and solid-state physics [33].

In optics, light beams composed of electromagnetic waves have a natural tendency to broaden as they propagate in a dispersive linear medium. Broadening of light can occur in space, in time or both. Pulses of light broaden in time due to the different frequency components of the temporal pulse travelling at different velocities (chromatic dispersion)[8]. In continuous wave light beams, spatial broadening is caused by diffraction, which can be understood by representing a beam as a linear superposition of plane-waves, where each one propagates at a slightly different angle and therefore at a different phase velocity with respect to the propagation axis [8]. In materials that possess significant optical nonlinearities, properties can be modified by the presence of light as reviewed in the previous Sections. In particular, when the medium modifies its refractive index with the presence of light, under certain conditions it is possible to counteract the spatial or temporal broadening obtaining a self-trapped beam or pulse. We will be mainly concerned with spatial self-trapping in optical beams. If we consider a Gaussian beam propagating along the z axis in a nonlinear medium, diffraction can be suppressed along z by a self-lens effect as seen in Figure 1.3. Here, diffraction creates a curve wavefront similar to a concave lens spreading the beam (Figure 1.3a). In a medium where the refractive index change is dependent on intensity, the change is largest at the beam center and gradually reduces to zero near the beam edges. This creates a self-focusing effect that acts similar to a convex lens focusing the beam towards the center (Figure 1.3b). In this way, light guides itself along the high-index region forming its own waveguide as it travels through the nonlinear medium. A balance between diffraction and self-focusing enables the beam to self-trap as shown in Figure 1.3c.

In the next Sections, we will first provide a brief review of self-trapping theory followed by self-trapping phenomena in various media. Then a review of previous experimental and theoretical studies of self-trapping in photopolymers will be presented along with some of its applications.

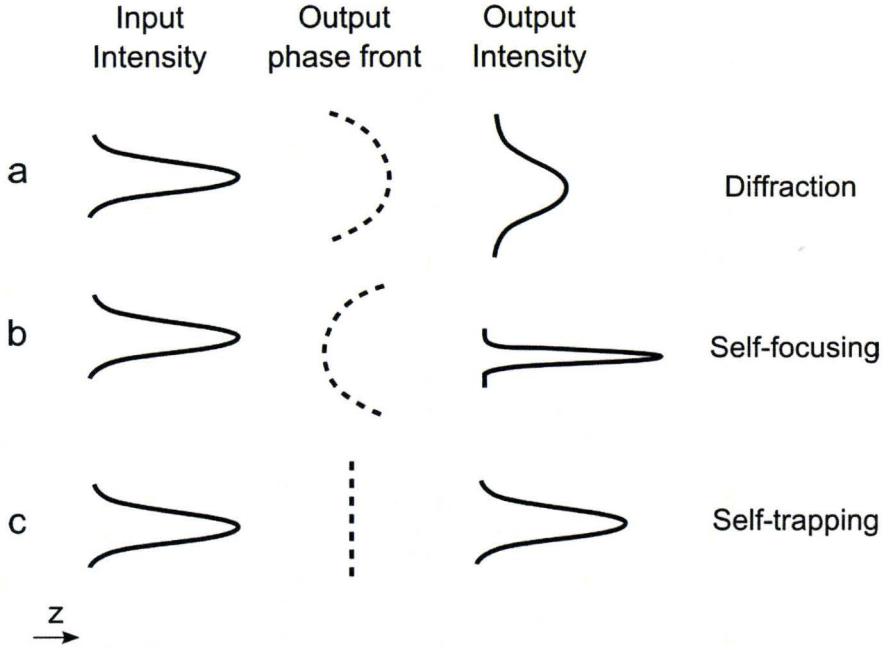


Figure 1.3: Scheme of optical self-trapping. Diffraction acts as a concave lens while the nonlinear medium acts as a convex lens. Self-trapping occurs when the two lenses balance each other and the phase front remains plane. Schematic illustration reprinted from [11] with permission from Elsevier .

1.2.1 Optical self-trapping theory

We will provide a brief overview of the main equations governing spatial self-trapping of light including the nonlinear parabolic equation and the nonlinear Schrödinger (NLS) equation, which is a special case describing self-trapping in Kerr nonlinearity. This last case is important as it marked the foundations of optical self-trapping where analytical solutions to the particular problem were possible.

Through Maxwell equations we can derive the following wave equation for the electric field E which generally describes the propagation of light in a medium [34]

$$\nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 P}{\partial t^2} \quad (1.8)$$

where c is the speed of light in vacuum and ϵ_0 is the vacuum permittivity. The induced polarization P consists of two parts

$$P(r, t) = P_L(r, t) + P_{NL}(r, t) \quad (1.9)$$

where P_L and P_{NL} are the linear and nonlinear part of the polarization, respectively. For spatial self-trapping we will focus on the case of a CW beam. A general solution of Equation (1.8) can be written in the form [11]

$$\mathbf{E}(r, t) = \frac{1}{2} \hat{x} [E(r, t) \exp(-i\omega_0 t) + c.c.] \quad (1.10)$$

where ω_0 is the carrier frequency, $\hat{x}$ is the polarization unit vector, and $E(r,t)$ is a slowly varying function of time. The polarization components P_L and P_{NL} can also be expressed in a similar manner. $E(r,t)$ can be expressed as

$$E(r,t) = A(r)\exp^{i\beta_0 z} \quad (1.11)$$

where $\beta_0 = k_0 n_0 \equiv 2\pi n_0/\lambda$ is the propagation constant in terms of the optical wavelength $\lambda = 2\pi c/\omega_0$. The beam propagates along the z axis and diffracts or self-focuses along the two transverse directions x and y , where x , y and z are the spatial coordinates associated with r . The function $A(x,y,z)$ describes the evolution of the beam envelope. When nonlinear and diffractive effects are included, and the slowly varying approximation (paraxial approximation) is applied to the envelope A so that $\partial^2 A/\partial z^2$ can be neglected, A is found to satisfy the nonlinear parabolic equation [11]

$$2i\beta_0 \frac{\partial A}{\partial z} + \left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} \right) + 2\beta_0 k_0 n_{nl}(I)A = 0 \quad (1.12)$$

where the first term represents the propagation of the beam envelope along z , the second and third term in parenthesis correspond to the diffraction of A in the transverse direction and the last term accounts for the nonlinearity in the medium, where $n_{nl}(I)$ is the nonlinear refractive index dependent on intensity. This is a general equation that can be used to model spatial self-trapping for various types of nonlinearities. In photopolymers, by substitution of $n_{nl}(I)$ with the appropriate refractive index change (Δn) dependent on intensity (Equation (1.7)), Equation (1.12) has been employed to model self-trapping [35].

For the case of Kerr nonlinearity where $n_{nl}(I) = n_2 I$, n_2 being the Kerr coefficient of the nonlinear medium, the following scaled dimensionless variables can be introduced [11]

$$X = x/w_0, \quad Y = y/w_0, \quad Z = z/L_d, \quad u = (k_0 | n_2 | L_d)^{1/2} A \quad (1.13)$$

where w_0 is a transverse scaling parameter related to the input beam width and $L_d = \beta_0 w_0^2$ is the Rayleigh range (or diffraction length). In terms of these dimensionless variables, Equation (1.12) can be expressed as the (2+1)-dimensional nonlinear Schrödinger equation [11]

$$i \frac{\partial u}{\partial Z} + \frac{1}{2} \left(\frac{\partial^2 u}{\partial X^2} + \frac{\partial^2 u}{\partial Y^2} \right) \pm |u|^2 u = 0 \quad (1.14)$$

where the sign depends on the sign of the nonlinear parameter n_2 . This equation was found to describe self-trapped beams in Kerr media by Chiao and coworkers in 1964 [10]. The (1+1)-dimensional version of Equation (1.14), which corresponds to the diffraction of the beam in only one transverse direction was solved analytically by Zakharov and Shabat [36] using the inverse-scattering method. Soliton solutions

were found corresponding to self-trapped beams where exact balance between diffraction and nonlinearity effects preserve the beam shape along the propagation direction. Unique properties of solitons were found solving the NLS equation including: integrability, conservation of power, velocity and number of solitons upon soliton collisions and fully elastic interactions amongst solitons. [11]. The distinction between solitons and self-trapped beams lies in that solitons are exact solutions of the NLS equation and therefore present the above properties that make their interactions similar to the ones of particles. While the term soliton was originally employed to describe self-trapped beams with the above properties, now they are sometimes used to refer to self-trapped beams in general.

1.2.2 Optical self-trapping and spatial solitons in various media

Optical spatial self-trapped beams have been observed in a variety of nonlinear media including Kerr media [3], photorefractives [37], photosensitive glasses [38], liquid crystals [14] and photopolymers [39]. The requirements for self-trapped beams, their dynamics and properties depend on the type of nonlinearity of the medium. As detailed in Section 1.1 the nonlinear photoresponse giving rise to refractive index changes can originate from high order susceptibilities as in the case of Kerr and photorefractive media, from modifications in the structural composition in photosensitive glasses, through the Fredericksen transition in liquid crystals and by chemical reactions in photopolymers.

Kerr media

The first observation of self-lensing or self-focusing effect was made in glass through the Kerr effect [40]. This was soon followed by the theoretical framework of Chiao and coworkers [10] in which the nonlinear Schrödinger equation was found to describe self-trapped beams or solitons in Kerr media. A relevant property of Kerr solitons obtained by solving the NLS equation is that they are only stable in (1+1)D, i.e. one longitudinal dimension along which the beam propagates and one transverse dimension along which the beam diffracts or self-traps. When an additional transverse dimension is added into which the beam can diffract (2+1)D, the beam undergoes catastrophic self-focusing and eventually breaks up [41]. Early experiments in Kerr media showed this behaviour often resulting in damage of the material [40]. The first (1+1) soliton in Kerr media was observed in 1985 in liquid CS₂ by employing an interference grating to obtain only one transverse dimension of diffraction [42]. But until 1990, a true (1+1)D soliton was observed in Kerr media by Aitchison and coworkers in a single-mode glass slab waveguide [3]. This was followed by observations of interactions between spatial solitons where their elastic collision properties were confirmed [43; 44]. Additional requirements for self-trapping in Kerr media include input powers in the order of kilowatts which are achievable using ultra-short laser pulses on the order of femtoseconds [3; 7].

Saturable media

The first demonstration of (2+1)D self-trapping was performed in 1974 by Bjorkholm and Ashkin [45] in atomic sodium vapour, close to a resonant transition. They explained the achievement by the effects of having a saturable nonlinearity. The stability of (2+1)D self-trapped beams in saturable media was explained later by Snyder and coworkers [46] by first giving the general definition that a soliton or self-trapped beam forms when the 'pulse' changes the potential in a way such that the pulse itself is a bound solution of that potential. Following this definition, we can understand that circular beams are stable in saturable nonlinear media as this implies that there is a maximum value of the optically induced refractive index change Δn_{sat} . Similar to Kerr media, a saturable medium focuses light increasing its intensity. However as index changes cannot exceed Δn_{sat} , the induced lens eventually becomes wider instead of stronger and decreases its focusing power. In Kerr media, catastrophic collapse occurs as a result of not being able to stop the self-focusing process. Another consequence in saturable media is that since the induced waveguide becomes wider, it is able to guide more than one guided mode [11]. The discovery of (2+1)D self-trapping in saturable media opened up the opportunity to find new kinds of spatial solitons in saturable media including photorefractive solitons and quadratic solitons. They gave rise to new family of soliton interactions in 3D that were not possible before as well as a variety of other phenomena.

Photorefractive solitons were first predicted by Segev and coworkers in 1992 [47] and later demonstrated by Duree and coworkers [48]. The nonlinearity giving rise to photorefractive solitons is the second order susceptibility (χ^2) (See Section 1.1). One of the first and most studied photorefractive solitons is the photorefractive screening soliton. Its generation starts with a narrow light beam propagating in a photorefractive crystal across which a voltage has been applied (Figure 1.4). As a result, free charges are photo-excited generating an internal space charge field, E_{sc}^0 , which is stronger in the darker regions compared to the more illuminated ones. As the refractive index change Δn is proportional to E_{sc}^0 , when Δn is negative, a graded index waveguide is generated guiding its own beam. The dependence of Δn on intensity is given by [8] $\Delta n = (V/L)(n^3 r_{eff}/2)[1/(|E|^2 + I_{dark})]$ for a (1+1)D screening soliton, where r_{eff} is the relevant term of the electro-optic tensor, V is the applied voltage between electrodes separated by a distance L and I_{dark} is dark irradiance which is a material parameter proportional to the conductivity of the crystal in the dark.

Other types of photorefractive solitons have been found including quasi-steady-state solitons [48], photovoltaic solitons [49], photorefractive semiconductor solitons [50] and solitons in centrosymmetric photorefractive media [51]. There are two important common properties to all photorefractive solitons. First is the ability to generate solitons at powers on the order of μW [12] because Δn depends on the ratio of $|E|^2 + I_{dark}$ and I_{dark} is low in photorefractive materials. The second property is that the response of the material is wavelength dependent which allows for soliton and waveguide generation at low powers that can be used to guide powerful beams at

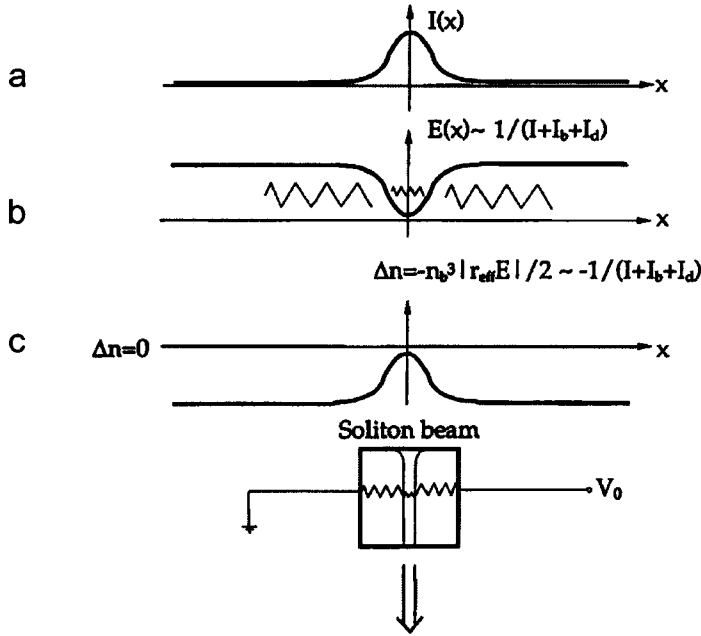
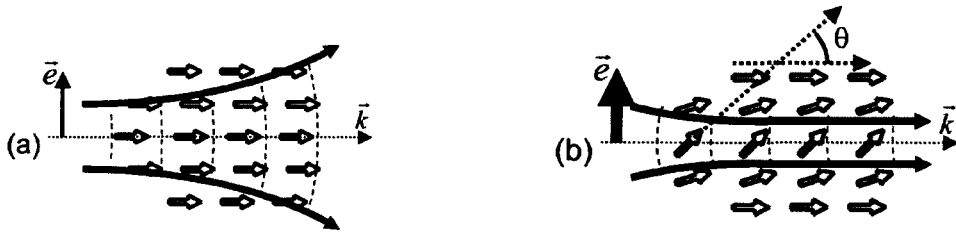


Figure 1.4: Formation of photorefractive screening soliton. After a) a non-uniform beam intensity distribution is input in a biased non-centrosymmetric crystal, b) a space charge field is internally formed resulting in c) a refractive index profile induced through Pockels effect which self-traps the beam. Scheme reprinted from [13] with permission from OSA.

wavelengths at which the material is less photosensitive [52]. Another type of soliton is the quadratic soliton which is generated in non-centrosymmetric materials in which phase matching is possible. They rely on second order nonlinearities $\chi^{(2)}$ and they result from the strong interaction and energy exchange between two or more beams at different frequencies possessing an effective saturable nonlinearity [17]. Solitons have been recently observed in nematic liquid crystals by Assanto and coworkers and have been called nematicons [15]. Their nonlinearity, as discussed in Section 1.1, originates from molecular reorientational processes. As seen in Figure 1.5, the beam behaves linearly at low intensities and at intensities above the Fredericks transition (or threshold intensity), the molecules orient themselves towards the electric field increasing the refractive index [15], resulting from a minimization in electric interaction energy density. Solitons can be generated in liquid crystals at mW power levels and over millimetre distances, they are saturable, non-local and substantially wavelength independent from visible to mid-infrared regions [14].

Interactions of solitons

Interactions of solitons have been studied and divided into coherent and incoherent interactions. The former occurs when the nonlinear medium can respond to interference effects between overlapping beams because the response time of the



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Figure 1.5: Scheme of optical self-trapping in liquid crystals [14]. The electric field is polarized ($\vec{e}$) perpendicularly to the molecular alignment ($\vec{k}$). a) Linear propagation where the beam diffracts and b) Self-focusing: the molecules are locally re-oriented, giving rise to a lensing effect along the beam axis.

medium is much smaller than changes in the relative phase of the beam. Therefore coherent interactions occur in nonlinearities with instantaneous time response such as Kerr materials and quadratic nonlinearity. [8]. Incoherent interactions refer to interactions where the relative phase between self-trapped beams varies much faster than the response time of the medium. In this case the medium only responds to the time-averaged intensity. Coherent interactions present attraction and repulsion of beams when these are in phase and out of phase (by π), respectively. Incoherent interactions only show attraction as the refractive index in between the beams always adds up due to the time-averaged intensity response.

In Kerr media, collisions are limited to only one plane as only (1+1)D solitons are stable. Since collisions are fully elastic, the number of solitons is always conserved. In contrast, in saturable media collisions are much richer. (2+1) D solitons are supported, therefore allowing for collisions in three dimensions. Self-induced waveguides in saturable media can guide more than one mode, which enables new phenomena including soliton fusion, fission and annihilation [53]. Spiraling-orbiting interaction has been demonstrated in photorefractive screening solitons [54] and nematic liquid crystals [14]. There, two solitons orbit and spiral about each other when the soliton attraction exactly balances the centrifugal force due to rotation as seen in Figure 1.6. These interactions between solitons have been proposed for the development of optical devices for all-optical signal routing, switching, steering and readdressing [15].

Other types of solitons

Other types of solitons that can exist in a variety of media are dark and vortex solitons [8]. The former consists of a narrow dark circular notch or stripe that propagates without broadening. Under linear conditions the notch broadens similar to a bright beam. Under nonlinear conditions, the refractive index decreases in the illuminated regions, creating a graded-index waveguide with high index in the dark region. This 'pulls' the background beam towards the notch reducing its diffraction. The resulting waveguide can then guide a bright beam. The amplitude of a dark soliton

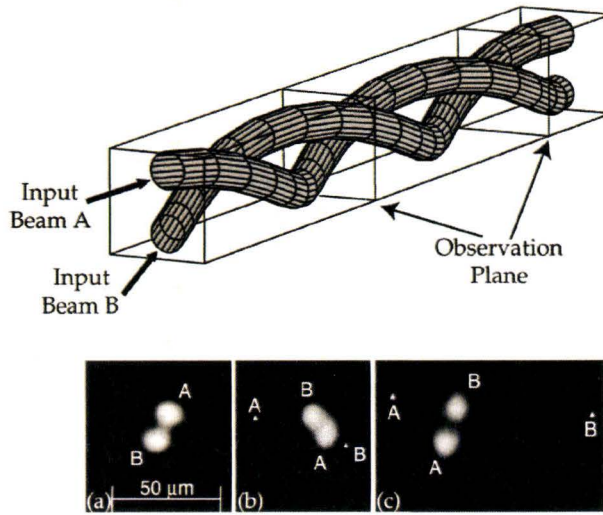


Figure 1.6: Illustration of soliton spiralling in photorefractive screening solitons launched at non-coplanar directions. a) Beams A and B at the input face of the crystal, b) the spiralling soliton pair after 6.5 mm of propagation, and c) after 13 mm of propagation. The centers of diffracting A and B are marked by white triangles. Illustration reprinted with permission from [54]. Copyright (1997) by the American Physical Society.

undergoes a π phase jump in the center of the notch. The two dimensional equivalent of dark solitons, i.e., (2+1)D, corresponds to vortex solitons. These are self-trapped beams with a phase front in the form of $\exp(im\theta)$, where θ is the azimuthal angle and m an integer referred as the topological charge.

Incoherent solitons are a relatively recent concept, until 1995 all soliton experiments required a coherent beam. That is, given a phase at a given location on the pulse in space or time it is possible to predict the phase in other locations. Demonstration of self-trapping of spatially and temporally incoherent white light beams has been performed in a slowly responding photorefractive crystal by Mitchell and Segev [55] and in a photopolymer by Zhang and coworkers [27]. Incoherent light is composed of speckles which result from random phase distribution which varies randomly with time. The main conditions for self-trapping of white light include a non-instantaneous nonlinearity with a response time much longer than the phase fluctuation time across the incoherent beam, the multimode beam should be able to induce a multimode waveguide and also be able to guide itself in its own induced waveguide.

1.2.3 Self-trapping studies in photopolymers and applications

The study of self-trapping in photopolymers is relatively recent, about 15 years, compare to its study in other nonlinear media such as Kerr and photorefractive ma-

terials. One distinguishing feature in photopolymers is that changes to the refractive index are permanent, therefore self-trapping of a beam inscribes a permanent waveguide structure. This feature has been the predominant subject of a large number of studies for potential applications of the self-written waveguides in integrated optics. Photopolymers also possess a saturable refractive index change that supports (2+1)D self-trapping, therefore rich dynamics and interactions between beams are expected to be observed.

In this section, a review of the previous experimental and theoretical work on self-trapping in photopolymers will be presented along with the studies on its potential applications.

1.2.3.1 Experimental studies

Experimental work on laser self-trapping due to free-radical polymerization was preceded by the demonstration of self-written tapered waveguides in UV-cured epoxy in 1993 by Frisken [56] using cw 532 nm light irradiated from an optical fiber, which allowed to modify the spot size of the fundamental mode of the fiber. Following, laser 2D self-trapping of photopolymers was demonstrated experimentally and through numerical simulations by Yariv and Kewitsch in 1996 [39; 20]; solid 10 micron-wide polymer fibers could be self-written with short exposures to a UV laser beam (325 nm) in a liquid diacrylate monomer.

Other studies on self-trapped beams have examined the filamentation, interaction between a pair of beams and *in situ* monitoring of polymerization. In experiments by Shoji and Kawata [57], photographs acquired along the propagation axis showed self-written fibres at 441.6 nm due to polymerisation in a urethane acrylate-based liquid resin; these experiments showed the formation of multiple self-trapped filaments with increasing optical power. Filamentation at increasing powers was also shown by Dorkenoo and coworkers, who carried out self-trapping experiments in 250 micron-thick layers of triacrylate-based resin [58; 59]. Shoji and coworkers investigated the interactions of a pair of self-trapped beams and their dependence on the collision angle and the relative power between the beams [60]. The temporal evolution of self-trapping with *in situ* Raman waveguide spectroscopy in planar waveguides was examined by Saravanamuttu and Andrews [61].

Self-trapping of incoherent (white) light and interactions of incoherent light self-trapped beams in photopolymers has recently been demonstrated in our group by Zhang and coworkers [27] and Kasala and Saravanamuttu [62].

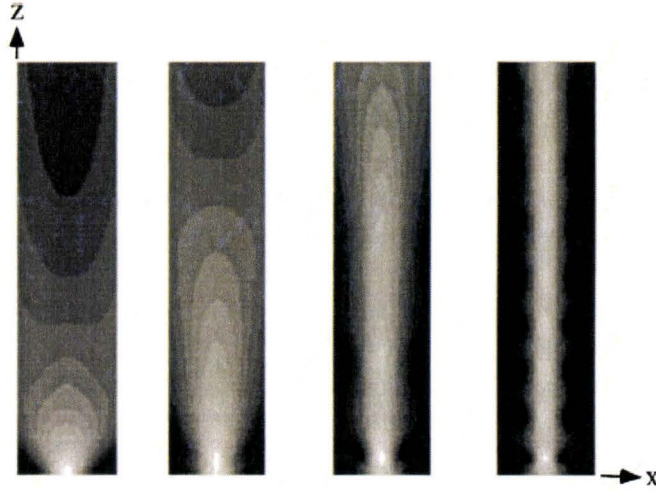


Figure 1.7: Numerical simulations of self-trapping using the beam propagation method by Kewitsch and Yariv. Illustration reprinted from [20] with permission from OSA.

1.2.3.2 Numerical studies

Self-trapping in photopolymers requires solving the nonlinear parabolic equation (Equation (1.12)) having a refractive index response corresponding to Equation (1.7). However, this is a complex mathematical problem and no analytical solution has been found yet, therefore numerical solutions are often proposed. The usual approach to solve the self-trapping problem in photopolymers is to solve Equation (1.12) numerically with updates in the refractive index changes following Equation (1.7).

Kewitsch and Yariv [20] were the first to investigate this problem in photopolymers and solved it numerically using the beam propagation method. The results in Figure 1.7 show the time evolution of a self-trapping beam, where the narrowing of the beam was evident over time as well as a high intensity area close to the entrance of the beam. Self-trapping was accompanied with width oscillations in the beam diameter.

Almost simultaneously Monro and coworkers [38] analysed numerically the self-trapping problem in (1+1)D in photosensitive media and in photopolymers, through the beam propagation method with updates in the refractive index changes. They also employed the Wentzel-Kramers-Brillouin [63] analysis to explain the modal properties of the induced waveguide. They found that as the refractive index increased with time, the number of modes also increased at every point in the propagation distance. The analysis included a self-written waveguide able to support two even modes. They found that as time progressed, the position at which the waveguide became multimoded changed, leading to a change in intensity distribution (secondary eyes) along the waveguide. Oscillations in intensity were found to be the result of mode beating. For that particular case, the beat length between the two even modes was found to be constant.

Other numerical studies have modelled self-trapping in photopolymers for an unpolymerized monomer [59] and by introducing an intensity threshold (I_{th}) [60]. In this last case, the waveguide width stabilized and did not present secondary eyes. Also a condition for single symmetric mode waveguide formation was proposed by having a maximum intensity (I_{max}) of the beam that followed $I_{th} < I_{max} < 7I_{th}$. In the same study, interactions of self-trapped beams in (1+1)D were numerically investigated.

1.2.3.3 Applications of self-induced waveguides

Emphasis has been placed on the circular fibers induced by self-trapped beams and their potential applications in integrated optics and fiber interconnect technology. Waveguides could be self-written in a photopolymerisable resin with 488 nm light emitted by multimode optical fibers [23; 64]. A coupling loss of 0.7db at $1.55\mu\text{m}$ in a 0.5mm gap between multimode fibers was achieved through the self-written waveguide [65]. Single-mode optical fibers have been shown to induce self-written waveguides with single mode propagation at 1310 nm creating fiber optical interconnects (Figure 1.8a) [66]. Yonemura and coworkers showed that a waveguide component of a wavelength division multiplexer could be self-written with light from an optical fiber (457 nm) (Figure 1.8b) [67]. Reconstruction of diced waveguides due to 45° mirror fabrication has been demonstrated through self-written waveguides [68]. Bachelot and coworkers showed the ability to fabricate micro-sized moulds of different intensity profiles at the tip of an optical fiber; by selectively propagating discrete higher order modes [69]. Arrays of micro-optical structures have been realized through self-trapping using masks [70; 71]. Another application of self-trapped beams is the fabrication of artificial ommatidia by Jeong and coworkers which mimics the imaging units of insect's compound eyes (Figure 1.8c) [72].

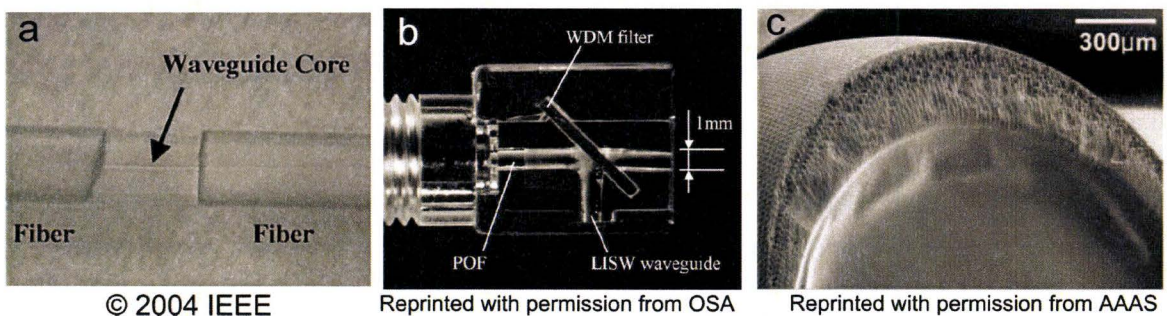


Figure 1.8: Applications of self-written waveguides in photopolymers in a) optical interconnects [66], b) a module for wavelength division multiplexing [67] and in c) fabrication of an artificial compound eye (ommatidium) [72].

1.3 Spatial self-phase modulation: diffraction rings

1.3.1 Introduction

Another spatial phenomenon observed in nonlinear optical media that undergo intensity-dependent refractive index changes is the generation of concentric rings of intensity with a Gaussian beam. Such diffraction rings emerge when the input beam suffers strong transversal modification of phase, which results in the interference of radiation with the same wavevector emanating from different points of the wavefront. Spatial self-phase modulation was first observed in media with thermally-dependent refractive index changes [73; 74; 75; 76; 4] and subsequently in atomic vapours,[5; 77] nematic liquid crystals [78; 6], Kerr media [75], chromophore-substituted silica [79] and photorefractive crystals [80].

Sections 1.3.2 and 1.3.3 provide an overview of the origin and requirements for the observation of diffraction rings in nonlinear media. Including the mechanism of spatial self-phase modulation and the role of input beam curvature in self-focusing and self-defocusing media in generating various ring patterns. A summary of diffraction rings studies in various nonlinear media and some of the potential applications of ring shape beams are also provided.

1.3.2 Origin of diffraction rings

In 1970, Dabby and coworkers [74] were the first to identify that diffraction rings originate from spatial self-phase modulation, which is the spatial analog of frequency broadening of short light pulses. In both cases, an intensity-dependent refractive index change modifies the phase of the pulse or beam of light resulting in the generation of new frequencies and in interference, respectively.

1.3.2.1 Spatial self-phase modulation

The spatial self-phase modulation mechanism is well established for thin media (thickness $<$ Rayleigh range) [6]. A laser beam that induces refractive index changes $\Delta n(r)$ as it propagates through a nonlinear material gives rise to a corresponding phase shift of the beam as given by

$$\Delta\psi(r) = \frac{2\pi}{\lambda} \int_{z_0}^{z_0+d} \Delta n(r, z) dz \quad (1.15)$$

where r is the direction that is transverse to the axis of beam propagation (z), z_0 is the position at the entrance face of the medium along z , d is the thickness of the

medium and λ , the wavelength of light in free space. In a medium where refractive index changes are intensity dependent, a beam with a TEM_{00} Gaussian intensity profile will induce a refractive index change with a similar profile. This in turn will induce a phase shift at the output of the medium expressed as

$$\Delta\psi(r) = \Delta\psi_0 \exp\left(-\frac{2r^2}{\omega^2}\right) \quad (1.16)$$

where ω is the beam radius at the entrance plane of the medium and $\Delta\psi_0$ is the maximum value of phase shift. In the phase shift profile, shown in Figure 1.9, specific points along r can have the same slope.

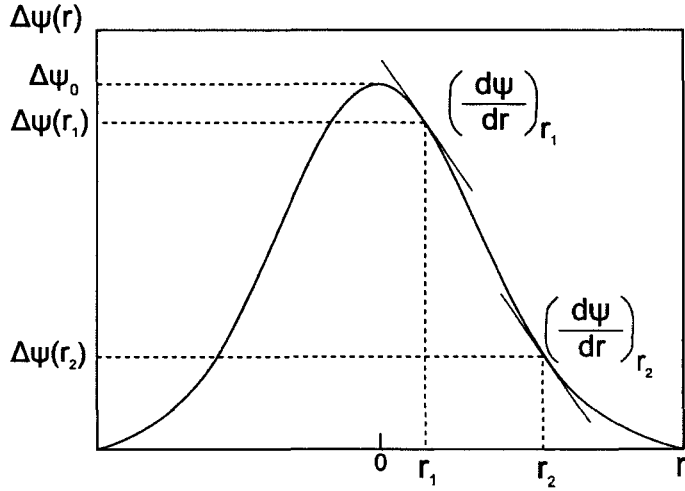


Figure 1.9: Schematic of the phase shift profile induced by a TEM_{00} Gaussian intensity profile in a nonlinear medium. Light emanating from points r_1 and r_2 have the same wavevector and can therefore interfere. Figure reprinted from [6] with permission of OSA.

The slope at each point in this curve corresponds to the perpendicular propagation wavevector $d\Delta\psi/dr = k_{\perp}$. Radiation from points with the same wavevector can therefore undergo interference. The condition for maximum constructive and destructive interference is given by $\Delta\psi(r_1) - \Delta\psi(r_2) = m\pi$, when m is an even and odd integer, respectively. Due to the cylindrical symmetry of the phase shift profile, interference results in concentric diffraction rings observed when projected onto a plane in the far-field. Multiple diffraction rings appear when $\Delta\psi_0 > 2\pi$; the number of rings N is estimated by

$$N \approx \frac{\Delta\psi_0}{2\pi} \quad (1.17)$$

Radiation from the inflection point of the $\Delta\psi(r)$ curve corresponds to the outermost ring. Its half-cone angle β can be estimated by [6]

$$\beta = \left(\frac{d\Delta\psi}{dr} \right)_{max} / (2\pi/\lambda) \quad (1.18)$$

In order to prevent simultaneous self-focusing of the beam in the nonlinear medium, the majority of studies of diffraction rings had to be carried out in thin samples (pathlengths $<$ Rayleigh length). For the case of thick media (pathlengths $>$ Rayleigh range), the description of the mechanism is more complex and not well understood yet for most materials. A more rigorous mathematical treatment is needed for each specific nonlinear mechanism to understand the effective thickness contributing to the spatial self-phase modulation. A study performed in strongly absorptive media, specifically sodium vapour [81], showed that two cases were possible in a thick sample. In the first, significant refractive index changes and therefore self-phase modulation only occurred near the entrance face of the medium. the resulting diffraction rings therefore underwent linear divergence through the remaining regions of the medium. In the second case, refractive index changes were dominant all along the propagation path, which led resulting to self-focusing and self-trapping of the beam.

1.3.3 Diffraction ring patterns

Different forms of diffraction rings can be elicited in nonlinear media, the most common being rings with dark or a bright central spot [82; 83]. The underlying mechanism for these pattern variations in patterns was developed by Santamanto [82], Yu [84] and Deng and coworkers [85]. The type of ring pattern depends on two factors, (i) the curvature of the beam (convergent or divergent) at the input face of the medium and (ii) the type of nonlinearity (focusing or defocusing). Self-focusing media exhibit positive refractive index changes whereas self-defocusing media exhibit negative refractive index changes with intensity [85; 76]. The theory developed by Deng and coworkers [85] successfully predicts ring patterns for four different scenarios and is briefly summarized here. The complex amplitude of a TEM_{00} Gaussian beam launched at the entrance of the nonlinear medium with thickness L is given by [85]

$$E(r, z_0) = E(0, z_0) \exp \left(-\frac{r^2}{\omega^2} \right) \exp \left(-\frac{ik_0 n_0 r^2}{2R} \right) \quad (1.19)$$

where k_0 and n_0 are the wavenumber and refractive index in free-space, respectively, ω is the beam radius at the entrance plane of the medium and R is the radius of wavefront curvature. Assuming that the medium is optically thin and the nonlinear absorption is negligible, the complex amplitude of the electric field on the exit plane of the medium can be written as [85]

$$E = (r, z_0 + L) = E(0, z_0) \exp \left(-\frac{\alpha L}{2} \right) \exp \left(-\frac{r^2}{\omega^2} \right) \exp(-i\psi(r)) \quad (1.20)$$

where α is the linear absorption coefficient of the nonlinear medium and $\psi(r)$, the total transversal phase shift induced by the beam at the exit plane of the medium can be written as

$$\psi(r) = \frac{k_0 n_0 r^2}{2R} + \Delta\psi(r) \quad (1.21)$$

Using Equation (1.16),

$$\psi(r) = \frac{k_0 n_0 r^2}{2R} + \Delta\psi_0 \exp\left(-\frac{2r^2}{\omega^2}\right) \quad (1.22)$$

where the first term corresponds to the contribution of the curvature R to the total transversal phase shift and the second term is the additional contribution from refractive index changes. The far-field intensity distribution of the beam at a distance D from the output face is obtained by applying the Fraunhofer approximation of the Fresnel-Kirchhoff diffraction formula on the optical field at the exit plane of the medium. The resulting far-field intensity is expressed as [85]

$$I = \left| \frac{1}{i\lambda D} \right|^2 \left| \int_0^\infty \int_0^{2\pi} E(0, z_0) \exp\left(-\frac{\alpha L}{2}\right) \times \exp(-ik_0 r \theta \cos\varphi) \exp\left[-\frac{r^2}{\omega^2} - i\psi(r)\right] r dr d\varphi \right|^2 \quad (1.23)$$

where θ is the far-field diffraction angle, φ is the angular coordinate on the exit plane of the medium in the polar coordinate system. The radial coordinate ρ in the far-field plane is related to the far-field diffraction angle θ by

$$\rho = D\theta \quad (1.24)$$

With the property of the first-kind zero-order Bessel function as an even function,

$$J_0(x) = \frac{1}{2\pi} \int_0^{2\pi} \exp(ix \cos\varphi) d\varphi = \frac{1}{2\pi} \int_0^{2\pi} \exp(-ix \cos\varphi) d\varphi \quad (1.25)$$

Equation (1.23) can be expressed as

$$I = I_0 \left| \int_0^\infty J_0(k_0 \theta r) \exp\left[-\frac{r^2}{\omega^2} - i\psi(r)\right] r dr \right|^2 \quad (1.26)$$

where $I_0 = 4\pi^2 |E(0, z_0) \exp(-\alpha L/2)/i\lambda D|^2$. Four cases are possible by combining convergent ($R < 0$) or divergent ($R > 0$) Gaussian beams propagating in self-focusing ($\Delta\psi > 0$) or self-defocusing media ($\Delta\psi < 0$). These are (i) $\Delta\psi(r) < 0$, $R > 0$, (ii) $\Delta\psi(r) > 0$, $R > 0$, (iii) $\Delta\psi(r) < 0$, $R < 0$ and (iv) $\Delta\psi(r) > 0$, $R < 0$. Since the sign of the total phase shift $\psi(r)$ does not change the result in Equation((1.26)), cases (i) and (iv) and cases (ii) and (iii) are equivalent. The resulting far-field intensity patterns for cases (i) and (iv) are concentric rings with a central dark spot and for cases (ii) and (iii) concentric rings with a bright central spot as shown in Figure 1.10.

1.3.4 Diffraction rings in various media and potential applications

The fundamental requirement for the generation of diffraction rings through self-phase modulation is a transversal phase shift ($\Delta\psi \geq 2\pi$) imposed on a Gaussian

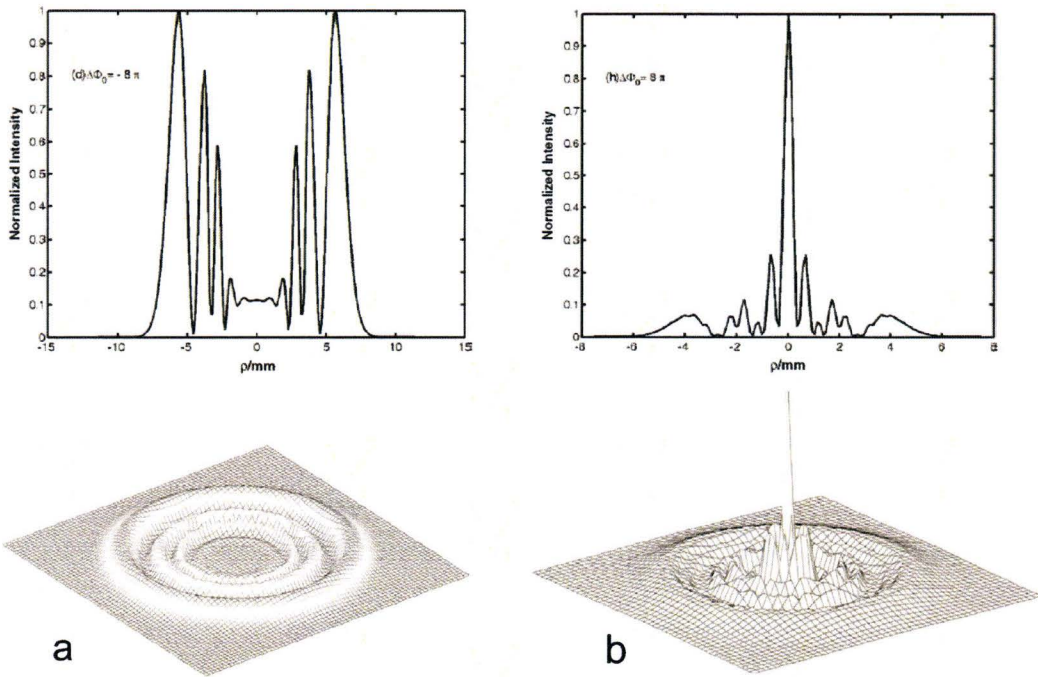


Figure 1.10: Simulation results from ring patterns for a) convergent ($R < 0$) and b) divergent ($R > 0$) input Gaussian beams in nonlinear media with self-focusing nonlinearity ($\Delta\psi > 0$). Images reprinted from [85] with permission from IOP.

beam. This requirement is satisfied by a variety of nonlinear media that undergo photoinduced changes in refractive index. Variations in the dynamics of diffraction rings originate from the photophysical and photochemical mechanisms that are unique to each nonlinear medium. For example, in the case of atomic vapours such as sodium [86], potassium [5] or barium [77] vapour, diffraction rings with frequency shifts with respect to the input frequency were observed using pulsed and cw lasers with sufficient intensity. This phenomenon was due to four wave mixing processes involving the third order susceptibility tensor $\chi^{(3)}$ [87]. The diffraction ring patterns were determined by the intensity of the beam, the detuning between the incident pulse and atomic resonance, and atomic density [88].

In nematic liquid crystals, diffraction rings were observed with cw lasers at input intensities above the Freedericksz transition, where birefringence is induced [6]. The number of diffraction rings increased with laser intensity due to the corresponding increase in phase shift imposed on the beam. Ring patterns with dark and bright central spots were observed in nematic liquid crystals for convergent and divergent beams respectively [82]. Diffraction rings in absorbing media originate from thermally-induced refractive index changes. Here absorption of light causes an intensity-dependent increase in the local temperature. The consequent intensity-dependent decrease in refractive index in regions of high intensity gives rise to a self-defocusing nonlinearity [73]. The number of rings was found to increase with beam power [4].

In most previous studies, diffraction rings were elicited in thin samples (path-lengths $<$ Rayleigh range) and observed in the far-field [6; 74; 4]. However, a recent study in a photorefractive crystal demonstrated that under certain conditions, diffraction rings could propagate over long distances ($\gg$ Rayleigh length) and were observed at the output of the nonlinear medium [80]. This provided an accessible nonlinear optical analogue to study the fundamental physics of dispersive shock waves that exist in superfluids such as Bose-Einstein condensates, which are considerably more challenging to generate experimentally.

Although diffraction rings generated in nonlinear media have not yet been directly used in any application, studies have explored their potential use as optical limiters [89; 90] and in super-resolving fluorescence microscopy [91] to improve the spatial resolution. Other potential applications that depend on ring-shaped beams have also been suggested including the measurement of thermal diffusivity in metal sheets by using a pulsed annular laser beam [92], as optical dark traps for atoms [93] and as optical tweezers for the manipulation of microscopic particles [94].

1.4 Filamentation due to modulation instability

The phenomenon of filamentation has been studied since the early 1960s. The first observations were made on liquid nonlinear media by Pilipetskii and Rustamov [95], where a laser beam at high intensities would break in filaments ordered in a random

manner. The first theory to explain filamentation of light beams was the modulation instability theory proposed by Bespalov and Talanov [96]. Filamentation has been observed in various media with coherent and incoherent light and also in beams with different shapes [97; 98; 99]. Particularly beams with ring shapes that develop during nonlinear propagation have recently shown its break up into multiple filaments [100; 99].

1.4.1 Modulation instability

Modulation instability in optical media has theoretically been studied mostly for Kerr media [101]. Bespalov and Talanov's modulational instability theory proposed that amplitude and phase perturbations in plane waves would develop and grow exponentially over the propagation path, until their strength was comparable to the initial plane-wave field, ultimately breaking up into multiple beams or filaments. The instability originated not only from small field variations or noise but also from random perturbations in the medium. To mathematically describe modulation instability in Kerr media, they introduced a perturbation e in the input electric field amplitude E [96]:

$$E = (E_0 + e)\exp(-i\gamma z) \quad (1.27)$$

where E_0 is the constant amplitude of the unperturbed wave, γ is the growth rate and $|e| \ll E_0$. By introducing Equation (1.27) in the nonlinear Schrödinger equation (Equation (1.14)), the theory predicted for plane waves characteristic scales of instability development that have dependence on input power and nonlinearity. Also relations for the power of an individual filament dependent on the input power were developed, an increasing number of filaments with increasing power of the initial beam and the reduction of power at which filamentation can occur when introducing ellipticity in the system.

Other instabilities that apply to self-trapped beams other than modulation instability in plane waves have been proposed as reviewed by Bergé and coworkers [102]. These include orbital stability, transverse instability and modulation instability on a ring. Orbital stability refers to the stability of an initial solution close to an equilibrium state to converge to a robust soliton shape. Transverse instability refers to a soliton mode perturbed by oscillatory fluctuations developing along one axis. Modulation instability on a ring refers to filamentation that occurs not uniformly over the entire beam but rather on rings developed after light propagation.

1.4.2 Observed filamentation in nonlinear media and its applications

Filamentation has been observed in self-focusing materials including Kerr media [95; 103], liquid crystals [104] and photopolymers [105; 106]. In Kerr media, the coalescence of filaments has been investigated with and without saturation [102].

Once filaments are created, they undergo self-focusing dynamics and their evolution is determined by the competition between the nonlinearity which confines the light in the filaments, its diffraction and the saturation of the nonlinearity. Fusion of filaments is dependent on their critical separation which in turn depends on their individual powers and whether or not saturation is present.

High-power femtosecond laser pulses in air have shown to filament and emit in a continuum from ultraviolet to infrared light due to the Kerr effect [107]. This white light emission filamentation shows an enhanced backward scattering detectable for over 20 km which can have applications in atmospheric trace-gas remote sensing [108]. Air ionization within the filaments holds promise for applications in laser-induced condensation and lightning control.

In photopolymers, filamentation has been theoretically and experimentally studied. Lonin and coworkers [109] found theoretically that the criterion for stable self-channeling regime (or no filamentation) is that the beam half-width be smaller than the sizes of the self-formed inhomogeneities ($\rho/a > 1$), where ρ corresponds to the radius of the inhomogeneities and a to the half-width of the input beam. This stable regime also works for broad beams at either lower radiation intensities or higher diffusion coefficients. However, self-channeling of a narrow beam can become unstable by increasing the light intensity or composite viscosity. They confirmed this experimentally at two different intensities corresponding to stable and non-stable regimes with laser (He-Ne) radiation experiments in a OKM-2 (oligocarbonate methacrylate) based photocomposite. Streppel and coworkers [105] have studied the effects of partially coherent light on modulation instability in photopolymers. They found that the threshold of minimum coherence length for modulation instability to exist depends only on systems parameters but not on light intensity. Spontaneous pattern formation has been observed due to modulation instability using incoherent white light in photopolymers with individual broad uniform beams [106] and during interactions of self-trapped white light beams [62]. 3D optical lattices with near cubic symmetry have been shown to form using interaction of orthogonal incoherent beams of light that simultaneously undergo modulation instability in a photopolymer [110].

1.4.3 Filamentation of ring-shaped beams

Filamentation of ring-shape profiles has been studied in nonlinear optics, mainly in Kerr media. These include optical vortices and rings formed during self-trapping having uniform phase. Optical vortices are beams with ring-shape profiles with a helical phase that have zero amplitude at the singularity where the phase is undefined and a topological charge m that is a measure of the phase winding [100]. Filamentation of optical vortices has been shown theoretically and experimentally under azimuthal modulation instabilities both in pure Kerr [100] and saturable Kerr-like media such as sodium vapor [111]. In a pure Kerr medium, filamentation was found to be a function of input power (P) and m . The analytical relation for number of azimuthal

filaments, η_{max} corresponds to $\eta_{max} = \sqrt{4P\alpha m^m / P_{cr}e^m - 2m - m^2}$, where P_{cr} is the critical power for vortex ring collapse and α is a constant dependent upon the initial beam shape [100]. In a saturable Kerr medium, the modulation instability of vortex rings depended only on the topological charge m and tended to break into $2m$ filaments [111]. Beams with uniform phase and ring shape also suffer filamentation during self-trapping due to modulation instability. For example, in carbon disulfide (a Kerr medium), ring diffraction patterns were formed by employing circular apertures before the entrance of the medium, these rings were observed to periodically breakup in the azimuthal direction due to instabilities [112]. In general in Kerr media, ring-shaped beams undergo azimuthal filamentation for sufficiently high powers, above the critical power for self-trapping [113; 114].

A related phenomenon is the self-trapping of necklace beams in Kerr media, these are beams shaped as rings whose radius is large compared to their radial thickness and their intensity is azimuthally periodically modulated, appearing as pearls or circular beams around a necklace [115]. These beams are different from filamentation of ring shape beams in that they do not filament but they are launched with a necklace shape. Necklace beams are the first type of beams described by the (2+1)D cubic self-focusing NLSE stable to transverse modulation instability. In order to arrest such instability in the azimuthal direction, the intensity is periodically modulated in the same direction. The main characteristic of necklace beams is that the radial dynamics rate of necklaces is many orders of magnitude slower than the rate at which each of the pearls of the necklace would suffer catastrophic collapse by itself. This results in necklace beams propagating stationary for large distances, during which neither the ring diameter nor the width of each pearl on the ring significantly change [115].

Recently super Gaussian (SG) input beams with uniform phase in Kerr media have been theoretically predicted [116] and experimentally observed [99] to self-trap into single ring-beams, which undergo filamentation due to non-azimuthal instabilities. By numerical integration of the (2+1)D NLSE (Equation (1.14)) for an initial beam with a uniform phase SG field distribution two regimes were found by Grow and coworkers [99]. At low powers of few times the critical powers (P_{cr}), the beam collapsed to a circular profile (or Townes profile) characteristic of Gaussian beams. However, at powers $\gg P_{cr}$ for self-trapping, the input SG beam collapsed into a ring-shape beam called the G-profile [116]. In this case, the initial formation of the ring could be understood by nonlinearity dominating over diffraction and the NLSE was approximated by [99]

$$i \frac{\partial u}{\partial Z} + |u|^2 u = 0 \quad (1.28)$$

where $u(X, Y, Z) = (r_0^2 k_0^2 n_2 c^3 / \pi)^{1/2} A(x, y, z)$, A is the amplitude of the envelope of the electric field, $X = x/r_0$, $Y = y/r_0$, $Z = z/2L_{df}$ and $L_{df} = k_0 r_0^2$ is the diffraction length, r_0 the characteristic radius of the input beam, $k_0 = 2\pi n_0 / \lambda$ is the wave number, λ is the vacuum wavelength, n_0 is the linear index of refraction, and n_2 is

the nonlinear index coefficient. The solution of Equation (1.28) for an input SG beam is given by [99]

$$u = u_0(X, Y)e^{ik_0 S} \quad (1.29)$$

where

$$S(Z) = \frac{n_2}{n_0} |u_0|^2 Z \quad (1.30)$$

For high powers the formation of an initial ring can be understood as the result of non-linear propagation over distances in which diffraction is negligible. The resulting ring radius increases with input power and the beam suffers filamentation in the presence of noise due to non-radially symmetric noise (noise non-symmetric around the central axis) at much lower powers ($P \sim 10P_{cr}$) than with a Gaussian beam ($P > \approx 100P_{cr}$). Through an azimuthal modulation instability analysis, an expression for the number of filaments, η , in super-Gaussian beams was found to be [99]

$$\eta = \sqrt{2e^{-1}(2Pw^2\alpha - e)} \quad (1.31)$$

where w determines the radius of the ring. The number of filaments according to Equation (1.31) is proportional to the square-root of the input power P .

1.5 Research objectives: laser induced self-action effects in photopolymers

As we reviewed in previous Sections, a variety of self-action effects have been predicted and observed in various nonlinear media. The specific dynamics for each phenomena and the interactions observed are strongly dependent on the type of non-linearity. In photopolymers, only self-trapping and filamentation of the beam have been demonstrated with the majority of studies having an emphasis on self-trapping and the applications of the resulting self-induced waveguides. However, theoretical studies in self-trapping have predicted complex dynamics of the beam evolution in photopolymers including excitation of high-order optical modes, complex evolution of the modal composition of the self-induced waveguide and oscillations of the self-trapped beam.

In this thesis, we aim to perform a systematic study in photopolymers to address the theoretical predictions described above and the lack of experimental investigations on the dynamics of self-trapped beams. These include the evolution from single mode to multimoded guiding during self-trapping, the sequential excitation of high-order optical modes and the correlation between oscillations in intensity and beam width and the modal composition of the self-induced waveguides. In order to do that, we performed an intensity dependent study as the optical photoresponse in photopolymers depends on intensity, that is photopolymerization rates and hence refractive index changes increase with intensity. This opened the opportunity to analyze the variation of self-trapping dynamics under increasing amounts of nonlinearity. The intensity dependence study performed covered 10 orders of magnitude,

which is the first study to our knowledge covering such a wide intensity range in a photopolymer. Moreover, a broader motivation to perform this wide range intensity dependence study was to acquire a better depth of knowledge of self-action effects in photopolymers. Unlike other optical materials such as photorefractive crystals, nonlinear propagation of light in photopolymers is in its infancy. Therefore, our aim was also to probe new forms of nonlinear light propagation. The experimental methods employed in this thesis differ from previous investigations in photopolymers, in that we employed beam profiling methods to directly visualize and quantitatively measure temporal and spatial changes in the cross sectional intensity profiles of the beam. As the response time of photopolymers is in the order of milliseconds to seconds [39], this was a feasible approach. Previous studies have only characterized self-trapping and filamentation through the light scattered along the beam path and properties of self-induced structures. Numerical simulations were also performed in this study in order to complement experimental findings and have a better insight into the observed phenomena. The study of self-action effects and dynamics of the nonlinear phenomena in the organosiloxane present potential not only for new discoveries and advances in photopolymers but also for a better understanding of self-action phenomena in other optical media. Moreover, new self-action effects in photopolymers are promising for optical applications as induced optical structures are permanent.

In this thesis first we present in Chapter 2, the materials and experimental methods employed for the experimental and numerical studies of self-action phenomena in the organosiloxane. Regarding experimental studies, preparation of the photosensitive organosiloxane and the optical assembly employed for beam profiling methods are described in detail. Conditions for monitoring the intensity profiles at different input intensities and the method to characterise resulting self-induced structures in our system are detailed. As for simulations studies, a description of the simulation procedure is provided. This include the beam propagation method used by BeamPROPTM software along with the use of the empirical formula (Equation (1.7)) to calculate refractive index changes suffered by the photopolymer. Physical and numerical parameters relevant to simulations are described and a simulation example is given at the end of that Chapter. This is followed by the intensity dependent study across 10 orders of magnitude in Chapter 3, where three intensity regimes are found. At low powers new self-trapping dynamics are experimentally confirmed including the excitation of high-order modes and its correlation with oscillation in beam intensity and width. At the mid-intensity and high intensity regimes novel self-action effects occur: diffraction rings due to self-phase modulation and single ring formation followed by filamentation, respectively. The origin of the different photoinduced refractive index profiles is examined and their importance to elicit the different optical phenomena is shown. Quantitative trends of self-trapping in the organosiloxane are presented through three important parameters: self-focusing time, self-trapped diameter and transmittance. Spatial profiles at all intensities are presented and analysed to illustrate the different phenomena at different intensity regimes. Chapter 4 is dedicated to study in depth the novel effect observed at the mid-intensity regime: diffraction

rings. Its origin is explained by linking the requirements for self-phase modulation and refractive index changes in the organosiloxane due to laser initiated free-radical polymerization. Dynamics of diffraction rings occurring in the organosiloxane at long pathlengths ($\gg$ Rayleigh range) are contrasted with previous studies mostly performed in short pathlengths. The temporal evolution of diffraction rings is presented and important parameters are extracted to relate the dynamics of diffraction ring formation with rates of polymerisation and refractive index profiles induced. The effect of beam curvature and pathlength dependence is examined showing the emergence of different types of diffraction rings and the complex dynamics occurring in a long pathlength medium. In Chapter 5, the next novel phenomenon is presented: formation of a single-ring from an input Gaussian beam and its subsequent spontaneous filamentation. We show that this effect is possible because of the flattening of the Gaussian refractive index profile induced, which originates from the saturation of the refractive index in the organosiloxane. The temporal evolution of the single-ring formation is shown overtime and its size and filamentation dependence on intensity is investigated. Permanent self-inscribed structures resulting from different nonlinear effects in the organosiloxane are shown through the thesis. Chapter 6 is dedicated to the study of 2D numerical simulations of the self-action effects in the organosiloxane found at a similar range of intensities studied in previous Chapters. This was achieved by employing the beam propagation method through BeamPROPTM software and an external subroutine which calculated the refractive index changes with Kewitsch's empirical formula (Equation (1.7)). Temporal evolution of intensity and refractive index profiles along the pathlength was simulated to be able to compare it with experimental results. Additionally, intensity profiles at the output of the medium were obtained, these profiles can be more directly compared to profiles obtained in experimental studies. Finally we summarize our main contributions of this thesis in Chapter 7 and propose potential studies for future work.

1.6 Published contributions to the field of nonlinear propagation of light in photopolymers

Contributions to the field of nonlinear propagation of light in photopolymers by the work performed in this thesis include: (i) systematic experimental study across 10 orders of magnitude in intensity in a photopolymerisable medium, (ii) observation of dynamics of nonlinear propagation in the organosiloxane through spatial and temporal monitoring of the beam profile, (iii) experimental confirmation of theoretical predictions in photopolymers including the transition from single moded to multimoded guidance of the self-trapped beam, the excitation of high order modes and its correlation with oscillations in beam intensity and width, (iv) identification of three intensity regimes in the organosiloxane eliciting novel phenomena: at low intensity self-trapping of the beam and excitation of high order modes, at mid-intensities the emergence of diffraction rings due to self-phase modulation and at high intensity regimes single ring

formation and its filamentation and filamentation of the whole beam, (v) importance of the refractive index profile induced to elicit the different observed phenomena, (vi) temporal dynamics of diffraction rings for long pathlength ($\gg$ Rayleigh range), (vii) effect of beam curvature and pathlength dependence on diffraction ring types and dynamics of diffraction ring propagation, respectively, (viii) intensity dependence of single-ring formation and its subsequent filamentation, (ix) permanent self-induced structures at the different intensity regimes and (x) 2D simulations of nonlinear light propagation in the organosiloxane at a wide range of intensities.

The published work include two peer reviewed articles, one conference proceedings and seven conference presentations:

Peer reviewed articles

- A. B. Villafranca and K. Saravanamuttu. An experimental study of the dynamics and temporal evolution of Self-Trapped laser beams in a photopolymerisable organosiloxane. *Journal of Physical Chemistry C*, 112(44):17388-17396, 2008.
- A. B. Villafranca and K. Saravanamuttu. Diffraction rings due to spatial self-phase modulation in a photopolymerisable medium. *Journal of Optics A: Pure and Applied Optics* 11:125202 (2009).

Conference proceedings

- A. B. Villafranca and K. Saravanamuttu. Nonlinear forms of laser propagation in a photopolymerisable medium. *SPIE Proceedings*, San Jose, CA, 7213:72130T (2009).

Conference presentations

- A. B. Villafranca and K. Saravanamuttu. Self-action effects of laser propagation in a photopolymer. *IV Confederation of Mexican Graduate Students and Researchers in Canada Congress*. Ottawa, ON, Canada (2009) [Oral presentation]
- A. B. Villafranca and K. Saravanamuttu. Laser-induced self-action phenomena in a photopolymerisable medium. *92nd Canadian Chemistry Conference and Exhibition-Canadian Society for Chemistry*. Hamilton, ON, Canada (2009) [Poster presentation]
- A. B. Villafranca and K. Saravanamuttu. Laser-induced self-action phenomena in a photopolymerisable medium. *Photonics North 2009*. Quebec, City, Canada (2009) [Poster presentation]

- A. B. Villafranca and K. Saravanamuttu. Laser-induced nonlinear phenomena in a photopolymer. *Women in Science and Engineering Initiative 2009 International Women's Day Research Conference*. Hamilton, Canada (2009) [Oral presentation]
- A. B. Villafranca and K. Saravanamuttu. Nonlinear forms of laser propagation in a photopolymerisable medium. *Photonics West 2009, Symposium on Integrated Optoelectronic Devices. Conference on Organic Photonic Materials and Devices XI*. San Jose CA, USA (2009) [Oral presentation]
- A. B. Villafranca and K. Saravanamuttu. Dynamics of self-trapped beams in a photopolymerisable medium: new opportunities for the design of self-written waveguide components. *TEXPO research competition, CMC Microsystems 2007 Annual Symposium*. Ottawa, Canada (2007) [Poster presentation]
- A. B. Villafranca and K. Saravanamuttu. Spatial self-trapping of coherent light in a photochemical system. *Photonics North, International Conference on Application of Photonic Technology, 2006*. Qubec City, Canada (2006) [Poster presentation]

Chapter 2

Materials and experimental methods

2.1 Introduction

In this Section, we describe the experimental and simulation procedures employed for studies of nonlinear light propagation in the organosiloxane photopolymer (Section 1.1.3). A detailed description of the preparation of the organosiloxane system is provided. The optical assembly and techniques used to characterise nonlinear light propagation and resulting microstructures in the organosiloxane are presented. We also provide numerical simulation procedures of nonlinear propagation of coherent light in the organosiloxane using BeamPROPTM and an external subroutine.

2.2 Preparation of photosensitive organosiloxane

Organosiloxanes were prepared through acid-catalyzed hydrolysis and condensation of 3-(trimethoxysilyl) propyl methacrylate (MAPTMS, Gelest, Inc., PA, USA). 1.1 g (5.5×10^{-5} mol) of 0.05 N hydrochloric acid (diluted from a standardized 0.1 N HCl aqueous solution, Sigma-Aldrich, Canada) was added to 17.6 g (0.0708 mol) of MAPTMS. The initially phase-separated mixture homogenized after 5 minutes of vigorous stirring to become a transparent colorless fluid. The sol was sensitized to visible light by addition of 0.05 wt.% of the free-radical photoinitiator (bis(η^5 cyclopentadienyl) bis(2,6-difluoro-3-(1H-pyrrol-yl)-phenyl) titanium(IV) ($\lambda_{max} = 393$ nm, 460 nm, Ciba Specialty Chemicals Inc., Canada) [21]. The sol was shielded from ambient light, stirred continuously for 6 days and filtered through a polytetrafluoroethylene (PTFE) membrane (0.2 μ m pore size, Pall Corporation, USA) prior to use. For self-trapping experiments, 1.8 ml of photosensitized sol was injected through a small perforation into a 6 mm-long home-made cylindrical cell with optically flat and transparent windows. The home-made cell consisted of microscope cover slips (25 x 25 mm) glued onto either side of a 6 mm-long plastic (Delrin) ring with an external diameter of 16 mm. The organosiloxane sol was then uniformly irradiated through one of the cell windows with white light from a quartz-tungsten-halogen lamp (440 s at 0.6 W, Cole-Parmer 09790-series, IL, USA) for approximately 10 minutes. In some experiments, a magnetic stir bar was incorporated into the home-made cell and illumination with white light was performed while the sol was stirred for 4 minutes. Gelation of the sol was indicated when the magnetic stir bar stopped

rotating. Irradiation caused partial polymerization of methacrylate groups, which in turn transformed the sol into a transparent orange gel that did not flow freely. To study the effect of pathlength on nonlinear light propagation, cells with pathlengths of 0.56, 2, 4, 8 and 10 mm were employed. The duration of irradiation varied for samples with different pathlengths and ranged from 2 to 15 minutes.

2.3 Optical assembly

Figure 2.1 is a scheme of the optical assembly constructed for self-trapping studies. The excitation source was the TEM_{00} mode (Gaussian beam $M^2 < 1.1$) of continuous wave, visible (532 nm) light emitted by a diode-pumped solid state laser (Verdi V5 Coherent, Inc., CA, USA). The output beam had a diameter and power of 2.25 mm and 100 mW, respectively. The beam was passed through a $\lambda/2$ wave plate (W1) and $\lambda/4$ wave plate (W2) and polarizing beam splitter cubes (C1 and C2), which were separated by an absorption filter (F). The wave plate orientations were adjusted to obtain the desired intensity for self-trapping studies. The attenuated beam was steered by two 45° elliptical mirrors (M) onto a planoconvex lens (L1, $f = 75.6$ mm), which focused the beam to a diameter of $20\ \mu\text{m}$ onto the entrance window of the cell containing the organosiloxane (S). The beam was linearly polarised in the y direction. The cell was mounted on a custom-made sample-holder, which could be translated along the optical axis (z) with a resolution of $0.5\ \mu\text{m}$. The cross-sectional (x , y) intensity profile of the beam at the exit face of the cell was imaged by a pair of planoconvex lenses (L2, $f = 100$ mm and L3, $f = 300$ mm) onto a high-resolution charge-coupled device (CCD) camera (736(H)x 484(V) pixels, pixel size $4.80\ \mu\text{m}$ (H) x $5.58\ \mu\text{m}$ (V); LaserCam IIID 1/4", Coherent Inc, CA, USA). The camera was driven by the BeamView Analyzer software (Version 3.2), which calculates beam diameter (FWHM, $1/e^2$), relative peak intensity, generates 2 and 3-D intensity profiles and compensates for image magnification by lenses L2, L3 (x 2.96). Other pairs of L1 and L2 lenses were used to change the magnification at the CCD camera. These include: L1, $f = 100$ mm and L2, $f = 75.6$ mm with a magnification of 0.74 and L1, $f = 62.9$ mm and L2, $f = 75.6$ mm with a magnification of 1.20. Combinations of absorption filters (F) mounted on three separate rotatable wheels (VARM, Coherent Inc.) were placed between imaging lenses L2 and L3 to prevent saturation of the CCD camera. All imaging optical components were mounted on carriers that could be translated along z with a resolution of 0.25 mm.

2.3.1 Divergence of laser beam under linear conditions

Measurements of the divergence of the beam under linear conditions were performed in order to calibrate the optical assembly. The divergence of the beam was monitored in air by measuring the beam diameter ($1/e^2$) at various distances away

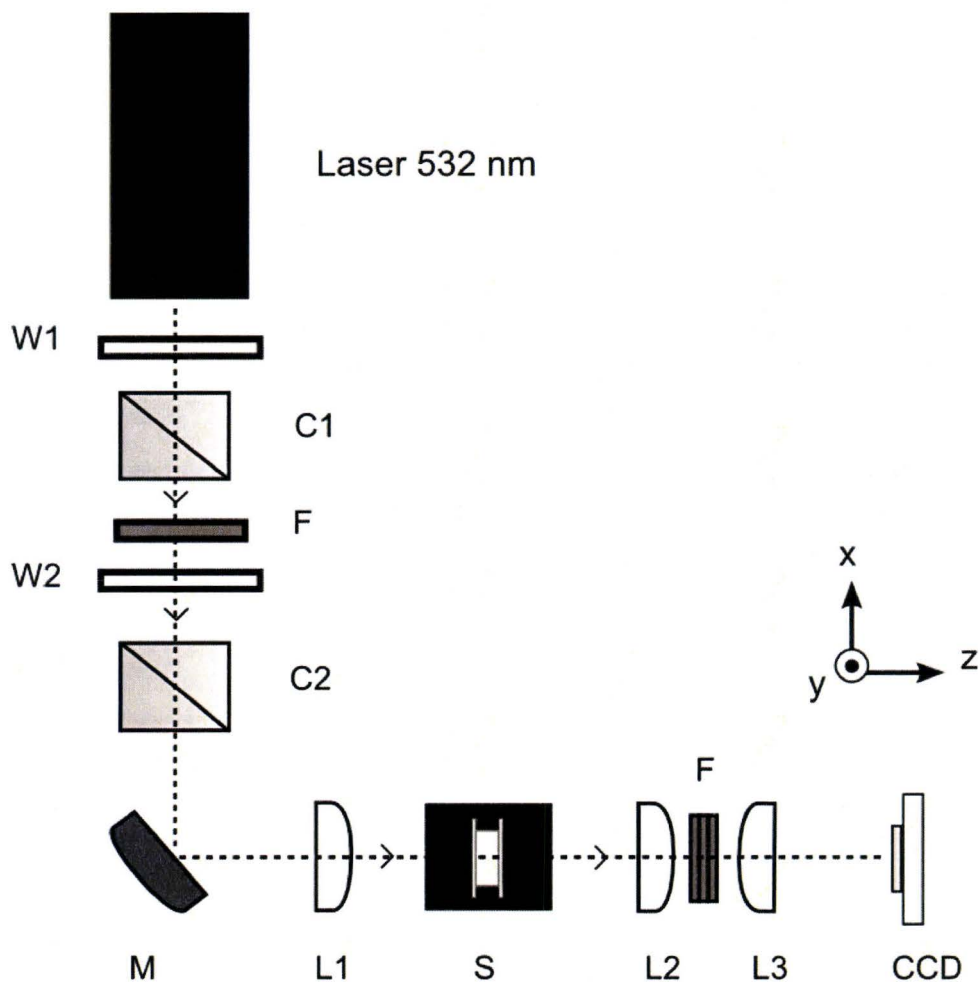


Figure 2.1: Optical assembly for nonlinear propagation studies in the organosiloxane. A 532 nm laser is attenuated with wave plates (W1 and W2), polarizing cubes (C1 and C2) and absorption filters (F). After reflecting from a mirror (M), the laser beam is focused with a lens (L1) onto the organosiloxane sample (S) and then imaged at the exit face with a couple of planoconvex lenses (L1 and L2) into a CCD camera (CCD).

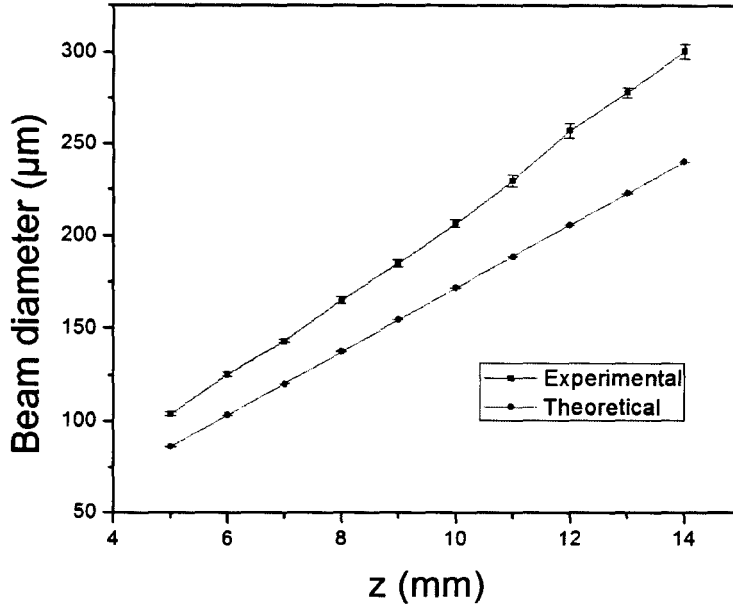


Figure 2.2: Laser beam divergence is shown for experimental and calculated values. The error lines for the experimental values reflect the standard deviation and for the calculated values the result of error propagation analysis.

from the focal point of the laser beam. The $1/e^2$ beam diameter was obtained through the *effective diameter* calculation included in the BeamView Analyzer software. This function calculates the diameter of a circle with an area equal to the area of all pixels with intensity above 13.5% of the measured beam peak intensity. The experimental beam divergence is plotted in Figure 2.2 together with its theoretical counterpart.

The theoretical calculations of laser beam divergence were performed by using the formula for the radius $\omega(z)$ of a Gaussian beam after certain propagation distance z [34]:

$$\omega(z) = \omega_0 \left[1 + \left(\frac{\lambda z}{\pi \omega_0^2} \right)^2 \right]^{1/2} \quad (2.1)$$

where ω_0 is the radius of the laser beam at the beam waist and λ is the wavelength of laser light. We calculated ω_0 with the approximated formula to obtain the radius of a Gaussian beam at the focal point after passing through a positive lens with focal length f [34]:

$$\omega_0 \cong \frac{f\lambda}{\pi\omega_l} \quad (2.2)$$

where ω_l is the radius at the beamwaist of the laser beam before passing through the positive lens. We obtained a theoretical radius after the lens of $\omega_0=9.9 \mu\text{m}$ by substitution of the experimental values $f=7.56 \text{ cm}$, $\lambda=532 \text{ nm}$ and $\omega_l=1.29 \text{ mm}$ in Equation 2.2. The corresponding experimental value $\omega_0=10 \mu\text{m}$ agreed well with the

theoretical calculation. However, the experimental and calculated divergence in air differs by an average of 22 % (Figure 2.2), which seems to be a systematic error. This error most likely originates from the inaccuracies in measuring the beam radius, which is then used to obtain the theoretical beam divergence of the beam. As the beam diameter is measured to be $20\text{ }\mu\text{m}$, but the pixel sizes in the horizontal and vertical direction are 4.80 and $5.58\text{ }\mu\text{m}$, respectively, magnification of the system is needed. To do that we magnified the image 2.96 times with the optical imaging lenses (L2, $f=100\text{ mm}$ and L3, $f=300\text{ mm}$), obtaining a beam diameter of $59.2\text{ }\mu\text{m}$. This improved our resolution, however further increase in magnification was not possible due to constraints with the optical set-up. Another possible source of error could be the positioning of the CCD in the z direction, which had a resolution of 0.25 mm .

2.3.2 Measurements at different intensities

In order to perform intensity dependent studies, the laser was tuned to the desired power and measured between the mirror and lense L1 in Figure 2.1. The measurement was taken with a power meter from Coherent Inc. consisting of a FieldMaster meter and the silicon detector LM-2-VIS, which is able to measure power from 10 nW to 50 mW in the wavelength range from 0.4 to $1.06\text{ }\mu\text{m}$. The intensity of the focused beam after L1 in Figure 2.1 was calculated by using $P/\pi r^2$ [34], where r was taken as the measured radius at the beam waist of the focused beam by the CCD camera.

The intensities calculated in our experiments are shown in Table 2.1 with the corresponding filters used to attenuate the light before it entered the CCD camera. The absorption filters labelled as w1, w2 and w3 correspond to the three filters labelled as F in the optical assembly shown in Figure 2.1.

All experiments were performed with the focused beam at the entrance face of the organosiloxane sample except in curvature dependence studies for diffraction rings for which the configuration is detailed in Section 4.6. The measurements performed at each intensity regime are summarised as follows:

Low intensity regime: The intensities covered for the low intensity regime include $3.2\times 10^{-5}\text{ W/cm}^2$, $1.6\times 10^{-4}\text{ W/cm}^2$, 0.003 W/cm^2 , 0.008 W/cm^2 and 0.016 W/cm^2 . The focused beam after L1 in the optical assembly of Figure 2.1 was placed at the entrance face of the sample for all intensities. The measurements made for all intensities include the temporal monitoring of the output beam at the exit face. For the intensity of 0.016 W/cm^2 , the propagation of light in the induced waveguide after self-trapping experiments was probed under linear conditions using an intensity of $3.2\times 10^{-5}\text{ W/cm}^2$. Preliminary multibeam experiments were performed also at 0.016 W/cm^2 by inserting a mask with a set of 7 circular apertures, before L1 in the optical assembly of Figure 2.1.

Mid-intensity regime: The intensities covered for the mid-intensity regime include 0.19 W/cm^2 , 1.6 W/cm^2 and 16 W/cm^2 . The focused beam after L1 in the optical

Table 2.1: Intensities employed with their corresponding filters

Intensity (W/cm ²)	Average power W	Absorption filters			
		transmittance w1	transmittance w2	transmittance w3	CCD filter (O.D.)
3.2x10 ⁻⁵	1x10 ⁻¹⁰	1.00	1.00	1.00	0.5
1.6x10 ⁻⁴	5x10 ⁻¹⁰	1.00	1.00	1.00	0.5
0.003	10x10 ⁻⁹	1.00	1.00	0.071	0.5
0.008	25x10 ⁻⁹	0.001	1.00	0.084	0.5
0.016	50x10 ⁻⁹	1.00	1.00	1.00	3.47(CCD filter)
0.19	600x10 ⁻⁹	1.00	1.00	1.00	3.47(CCD filter)
1.6	5x10 ⁻⁶	1	0.021	0.06	3.47(CCD filter)
16	50x10 ⁻⁶	0.001	0.035	0.06	3.47(CCD filter)
27	85x10 ⁻⁶	0.01	0.021	0.06	1.3
40	125x10 ⁻⁶	0.001	0.035	0.084	1.3
64	200x10 ⁻⁶	0.010	1.00	0.084	1.3
80	250x10 ⁻⁶	0.001	0.021	0.084	1.3
95	300x10 ⁻⁶	0.1	0.021	0.06	1.3
111	350x10 ⁻⁶	0.1	0.035	0.071	1.3
159	500x10 ⁻⁶	0.001	0.035	0.06	3.47(CCD filter)
1592	5x10 ⁻³	0.1	0.021	0.071	3.47(CCD filter)
12732	40x10 ⁻³	0.1	0.06	0.06	3.47(CCD filter)

assembly of Figure 2.1 was placed at the entrance face of the sample for all intensities. The measurements made for all intensities include the temporal monitoring of the output beam at the exit face. At an intensity of 1.6 W/cm², studies of the input beam curvature dependence were performed by placing the entrance face of the sample 2 mm to the right and to the left of the focal point. A path length dependent study was performed also at 1.6 W/cm², using samples with thicknesses of 0.56 mm, 2 mm, 4 mm, 6 mm, 8 mm and 10 mm. In these cases, imaging distances included the imaging at the output face and further away. The output intensity patterns of the 0.56 mm sample were taken with a Canon PowerShot SD1000 camera at a distance of ≈ 30 cm from the exit face. The output intensity patterns of the 2 mm sample were imaged at 0 mm, 2.5 mm, 10 mm, 13 mm, 18 mm, 19 mm and 20 mm from the exit face. For the 4 mm sample, the imaging distance was 0 mm, 2.5 mm and 10 mm from the exit face and for the 6mm sample, they were 0 mm, 2.5 mm, 9 mm and 12 mm from the exit face. The output intensity patterns of 8 mm and 10 mm samples were only imaged at the output face.

High intensity regime: The intensities covered for the high-intensity regime include 27 W/cm², 40 W/cm², 64 W/cm², 80 W/cm², 95 W/cm², 111 W/cm², 159 W/cm², 1592 W/cm² and 12732 W/cm². The focused beam after L1 in the optical assembly of Figure 2.1 was placed at the entrance face of the sample for all intensities. The measurements made for all intensities included the temporal monitoring of the output

beam at the exit face. Additional measurements for the individual size of filaments were performed with the CCD software (Beam View Analyzer) by using the function "inclusion". This function can calculate the beam diameter of a selected area of an image taken by the CCD camera.

2.4 Characterisation of self-induced structures in organosiloxane

Characterisation of self-inscribed structures due to nonlinear propagation was carried out through reflection and transmission optical microscopy (Olympus BX51 and SteREO Discovery.V12, maximum magnification 400x, Carl Zeiss Ltd.). Micrographs were acquired by inserting a red filter in the white light path to protect the sample from further polymerization. Micrographs were acquired with two configurations: under transmission, the entrance and exit face of the waveguide were aligned with the line of vision and under reflection, the waveguide length was almost perpendicular to the line of vision. In this case, micrographs were acquired by placing the sample into a rotatable holder with a reflective surface (aluminum foil) underneath and were taken at different angles to capture the full length of the structures.

Self-written waveguides were passively probed by coupling low intensity (3.2×10^{-5} W /cm²) 532 nm laser light. After the waveguide was induced, the laser intensity was tuned to the lower intensity without displacing the waveguide and images of the intensity profiles at the output face were recorded with the CCD camera using the same optical assembly in Figure 2.1. Polymerization at these intensities is extremely slow, enabling characterization of waveguides under passive conditions.

2.5 Numerical simulations

As detailed in Chapter 1, nonlinear light propagation, specifically self-trapping, has been theoretically described in various media using a variety of techniques [117; 11]. Solutions to the nonlinear Schrödinger (NLS) equation describe the self-trapping process in nonlinear media [117]. In photopolymers, the equivalent expression, the nonlinear paraxial wave equation has been numerically [35] and semi analytically [63] solved. The refractive index change is given by the empirical expression developed by Kewitsch and Yariv and given by [20]

$$\Delta n(x, y, z, t) = \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(t)|^2 dt \right] \right\} \quad (2.3)$$

where Δn_s is the maximum refractive index change, U_0 is the critical exposure required to initiate polymerization, τ is the monomer radical lifetime and $E(t)$ is the amplitude of the electric field.

In the case of other nonlinear phenomena such as the emergence of diffraction rings, modelling has generally included the diffraction patterns for the far field in thin optical media using the phase shift of the optical field after passing through it and the Fraunhofer approximation of the Fresnel-Kirchhoff diffraction formula [85].

We modeled 2D nonlinear propagation of a Gaussian beam in the organosiloxane through an iterative process using the beam propagation method (BPM) combined with calculation of updated refractive index changes in the medium. The BPM was used with the aid of the software package BeamPROPTM (RSoft Design Group, Inc) and the refractive index calculations through an external subroutine (Appendix B). Specifically, a Gaussian beam was launched in a uniform refractive index medium, its propagation under linear conditions was calculated and a map of the resulting electric field amplitude along the propagation length was obtained. Then using Equation (2.3) in the external subroutine, refractive index changes were calculated based on the electric field amplitude map and those changes were input in the BeamPROPTM software, where the propagation of the Gaussian beam through this modified medium was modelled again. This process was iteratively performed over many steps. This approach is equivalent to nonlinear propagation in the photopolymer because refractive index changes are permanent therefore by updating the new changes in refractive index in the medium, we take into account the nonlinearity of the system. Comparisons with results obtained by solving the nonlinear paraxial wave equation showed excellent agreement [63]. The BPM is an approximation of the exact wave equation for monochromatic waves. By using a scalar field assumption, the wave equation reduces to the Helmholtz equation and by using the slowly varying envelope approximation (paraxial approximation) the Helmholtz equation reduces to [118]

$$\frac{\partial u}{\partial z} = \frac{i}{2k} \left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + (k^2 - \bar{k}^2)u \right) \quad (2.4)$$

having

$$\phi(x, y, z) = u(x, y, z)e^{i\bar{k}z} \quad (2.5)$$

where $u(x, y, z)$ is the slowly varying field, $\bar{k}$ is a constant number that represents the average phase variation of the field $\phi(x, y, z)$, k is the wave number and x and y are the transverse coordinates and z is the propagation coordinate. We performed 2-D simulations in BeamPROPTM, which is a simplification of Equation (2.4), where the y dependence is omitted. This can be justified by the radial symmetry of the refractive index changes in the isotropic organosiloxane.

2.5.1 Simulation procedure

The steps for nonlinear simulations in the organosiloxane are as follows:

Step 1: A file with *.ind extension was generated in BeamPROP where a 20 μm diameter Gaussian beam with a wavelength of 532 nm was launched in the central

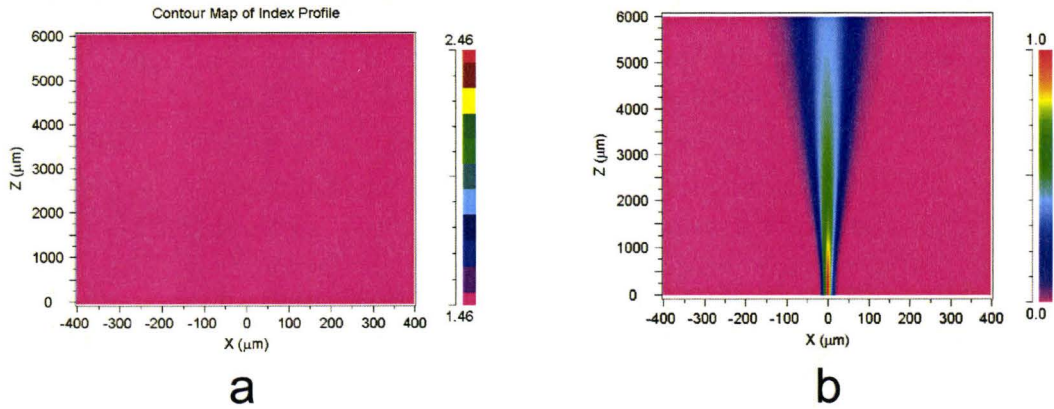


Figure 2.3: Step 1 of simulation where a Gaussian beam is launched into a) a block of uniform refractive index resulting in b) the 2D propagation of the electric field.

part of a 2D block ($800 \mu\text{m} \times 6.00 \text{ mm}$) with uniform refractive index (Figure 2.3a). These parameters corresponded to the experimental ones, except in the case of the width of the 2D block which was reduced to $800 \mu\text{m}$ in order to reduce computation time. The resulting electric field amplitude map shown in Figure 2.3b corresponded to the first step in the simulation.

Step 2: The electric field amplitude map was exported to the external subroutine (Appendix B) as slices of values across the propagation direction z in text format. The external subroutine combined all the slices into a map of electric field amplitudes and converted it into a map of intensity values. By employing Equation (2.3) for refractive index changes in photopolymers in the subroutine, the intensity map was converted into a refractive index map and exported in text format.

Step 3: A second file with *.ind extension was generated in BeamPROP where the refractive index map was imported, having now a 2D block ($800 \mu\text{m} \times 6.00 \text{ mm}$) with a refractive index profile as shown in Figure 2.4a. A Gaussian beam, with same characteristics as in Step 1, was launched into this new index profile and the simulation resulted in a new electric field amplitude map as shown in Figure 2.4b. After this step, we repeated step 2 continuously where each computational step represented time, therefore we were able to simulate changes over time in refractive index and the resulting propagating optical field.

The background refractive index in step 1 was set at 1.46 and the maximum refractive index Δn_s in Equation (2.3) used in step 2 was set at 0.006, this approximated value is based on previous measurements performed on thin films of the organosiloxane [24].

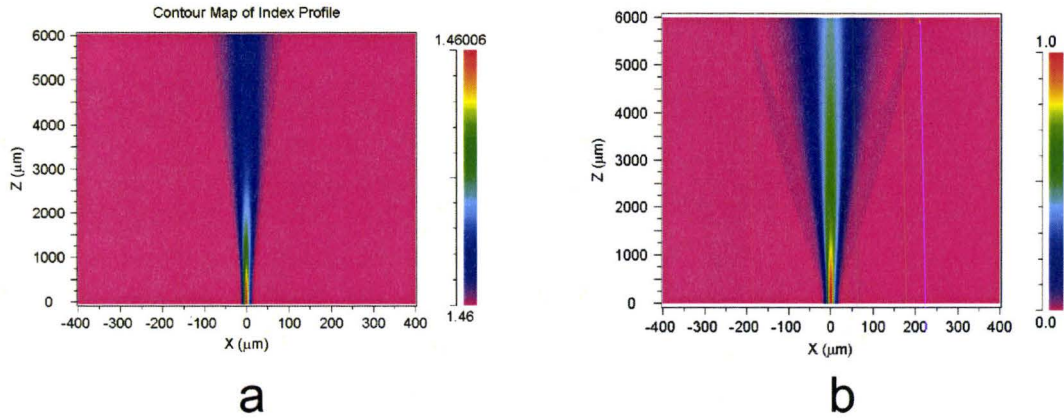


Figure 2.4: Step 3 of simulation where a Gaussian beam is launched into a) a block with a refractive index profile resulting in b) the 2D propagation of the electric field.

2.5.2 Simulation parameters

The main variables for numerical simulations can be separated in two groups: physical parameters and numerical parameters. Physical parameters correspond to those variables related to the physical process of refractive index changes due to photopolymerisation in the organosiloxane. These include the maximum refractive index change Δn_s and critical exposure U_0 (Equation (2.3)). Numerical parameters are variables that were used to optimize the simulations, these include: grid size and slice grid in x and z coordinates and intensity threshold.

Physical parameters

Δn_s is the maximum refractive index change possible in the organosiloxane systems and it is one of the variables in Equation (2.3) which we use in Step 2 to calculate refractive index changes at every spatial point.

Critical exposure (U_0) value is the exposure needed to induce polymerization as indicated in Equation (2.3). In simulations, we employed U_0 as a parameter to tune the response of the refractive index to intensity through the use of Equation (2.3) in the external subroutine. We used this approach because BeamPROPTM software does not allow for different input beam intensities. As we detail in Section 6.2, by having the physical critical exposure of the system U_0 , the refractive index saturates at different times depending on input intensity. However, in simulations with BeamPROPTM software, we cannot fix U_0 and vary the input intensity, instead we fix the input intensity and vary U_0 to achieve a similar response in the refractive index change which is our primary concern. As the input power is always one and has no units in BeamPROPTM software, we did not employ units for the parameter U_0 in simulations. Another reason for not including units in the parameter U_0 is to avoid confusion with the physical value of the system. In simulations changing this parameter does not mean that we have a new physical system with its particular critical

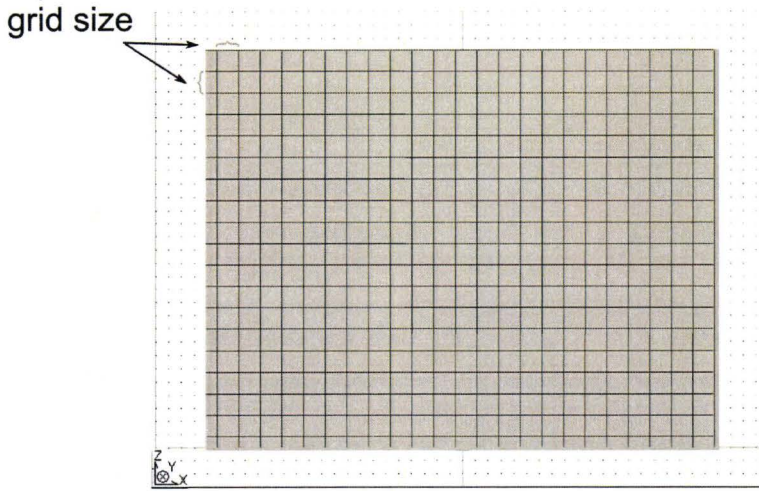


Figure 2.5: Scheme of simulation area in red showing the grid size in BeamPROPTM software

exposure U_0 but rather that we have the same system which saturates its refractive index at different times depending on the input intensity.

Numerical parameters

Grid size is the size of the mesh used to calculate the propagating field in BeamPROP. As shown in Figure 2.5 the parameter can be set for both x and z coordinates. In order to optimize the grid size, a convergence study was performed by monitoring the beam parameters: $1/e$ width and launch power of the beam along z . The launch power is the overlap integral between the calculated field at the current z position and the input field for the simulation. The convergence study consists on monitoring these parameters under identical launching and sample conditions but with varying grid size value. The convergence occurs at a particular grid size value at which the parameters do not change for the next lower value for grid size. At this point, even if the grid size is decreased, accuracy of the results will not improve but computational time will increase. The range of grid size values employed varied from 0.2 to $0.003 \mu\text{m}$ for values corresponding to $0.2 \mu\text{m}/(2^N)$ for $N=0, 1, 2, 3, 4, 5, 6$. The $1/e$ width and launch power values were monitored for all the calculated N 's. A grid size of $0.05 \mu\text{m}$ was found to be a compromise between computational time and accuracy in results. The variation of $1/e$ width and launch power compared to the most stable N value was $0.0005 \mu\text{m}$ ($3.6 \times 10^{-4} \%$) and 0.3×10^{-6} relative units of power ($1.07 \times 10^{-4} \%$), respectively.

Slice grid is the size of the mesh used to display results in BeamPROPTM as shown in Figure 2.6. This parameter is important because depending on its value, the visual resolution of the results can be adjusted. In our case, the slice grid was also important because this value determined the resolution of the exported electric

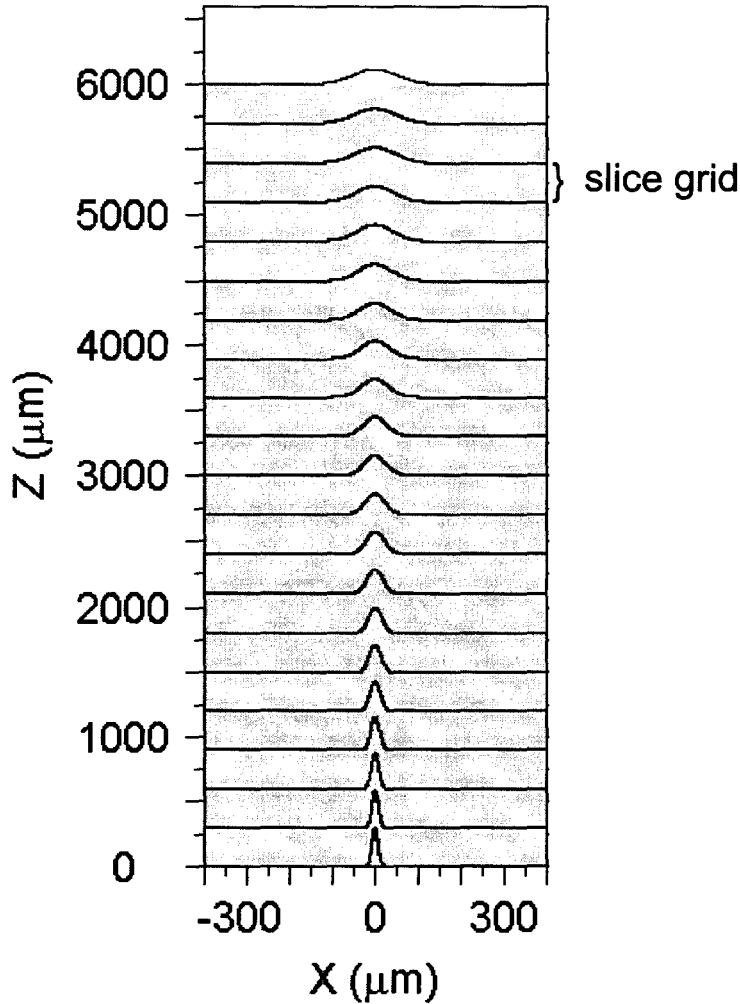


Figure 2.6: Resulting field after a simulation in BeamPROPTM software showing the slice grid

field amplitude values which determined the resolution of the refractive index map calculated and imported in every simulation step. This value was set to $0.4 \mu m$. In principle the smaller the value, the better the spatial resolution for the refractive index map, however, the external program (see Appendix B) had a limitation on the array size corresponding to these values (N_x and N_z in the program).

Intensity threshold is another parameter that was introduced in the external sub-routine (Appendix B) as the variable "thresh". This parameter sets an intensity filter, resulting in no refractive index changes for those regions where intensity values are below "thresh". This parameter helped to simulate cases where refractive index changes in outer parts of the optical field (below a certain intensity) were thought to be insignificant in real experiments.

2.5.3 Simulation example

Figure 2.7 shows a sequence of simulation steps showing self-trapping. At the top, the 2D refractive index profile corresponding to the initial block with uniform refractive index (1.46) is shown. In this case a Gaussian beam with $10\text{ }\mu\text{m}$ radius was launched into the uniform sample. Step 1 in column (a), shows the resulting 2D intensity profile after propagation of the Gaussian beam into the uniform block. We observe that the beam diverges as it propagates. Step 1 in column (b) shows the resulting 2D refractive index profile calculated by the external subroutine. This was done by using the field amplitude values from the intensity profile in Step 1 (column b) in Equation (2.3). The index profile is then imported BeamPropTM as the new index profile. The Gaussian beam is launched in the new index profile and its propagation is calculated. This process continues iteratively obtaining the propagation of the Gaussian beam over time. We see over 29 computational steps the narrowing of the beam (column a) and the resulting waveguide formation (column b).

In this particular example $U_0=500$ was employed. By varying this parameter in Equation (2.3) in the external subroutine we carried out simulations at different intensities. As detailed in Section 6.2, decreasing U_0 was equivalent to increase the intensity in our simulations.

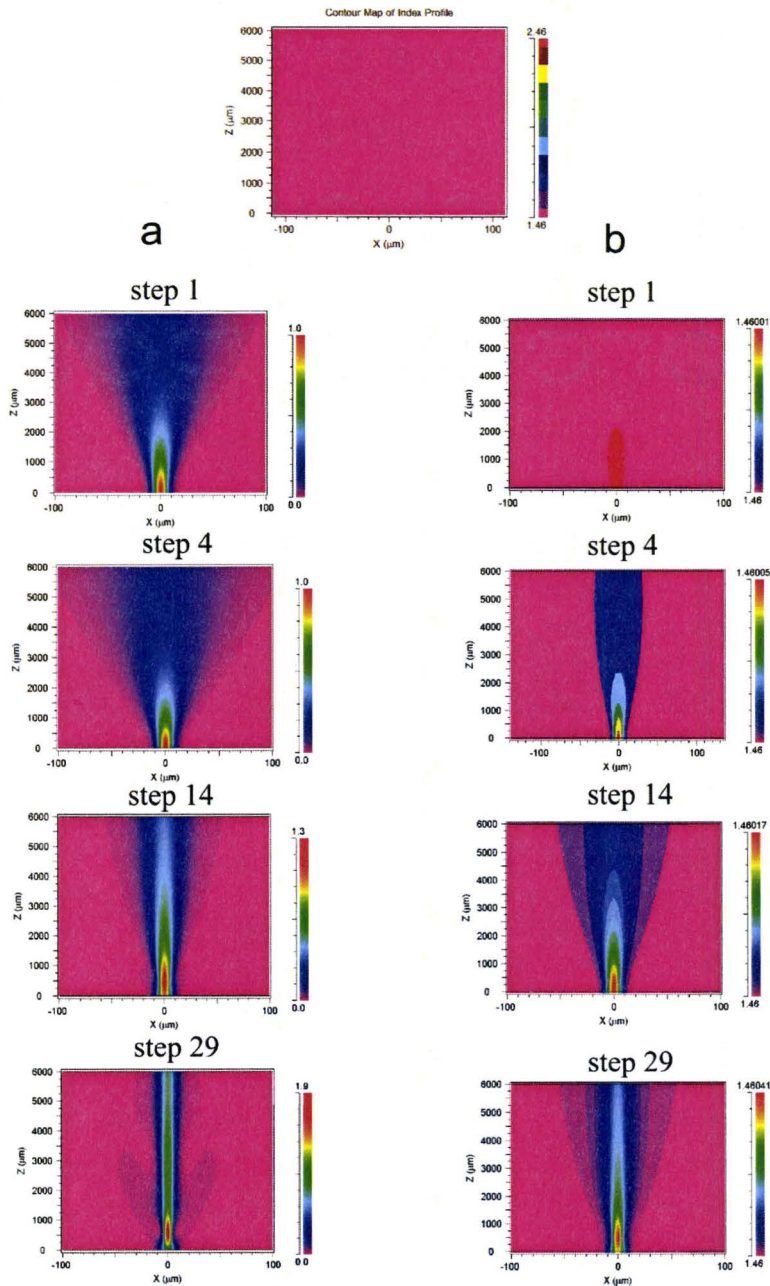


Figure 2.7: Sequence of simulation steps showing self-trapping in BeamPROP™. At the top, the 2D refractive index profile of the initial block with uniform refractive index (1.46) is shown. In a) 2D intensity profiles are shown for $U_0=500$ for selected steps and in b) the corresponding 2D refractive index profile are presented.

Chapter 3

Intensity dependence of nonlinear light propagation in organosiloxane

3.1 Introduction

The natural diffraction of light can be suppressed in a medium that exhibits photo-induced changes in refractive index. Under these conditions, a typically Gaussian beam becomes entrapped within a self-induced waveguide and propagates without broadening over distances $\gg$ Rayleigh range. Self-trapped beams are expressed as solutions of the nonlinear Schrödinger equation, which describes competition between the natural diffraction and self-induced refraction of the beam. Optical self-trapping has been studied across forty years [117] in materials as varied as Kerr media [10], photorefractive crystals [8], photosensitive glasses [35] and photopolymerizable resins [39; 20]. The dynamics and temporal evolution of self-trapped beams however are determined by the molecular-origins of refractive index changes in the medium [117; 53]. Striking differences exist for example between self-trapping dynamics in the two most frequently employed materials, Kerr media and photorefractive crystals. Because refractive index changes in the former originate from the third-order susceptibility tensor, self-trapping can only be elicited with intense (GW/cm^2 to TW/cm^2), short (10^{-15} s) pulses of light. The non-saturable nature of the Kerr response moreover renders 2-D self-trapped beams unstable, restricting experiments to planar waveguide (1-D) configurations. While early studies of 1-D self-trapping in Kerr media were seminal and provided elegant mathematical solutions [10], more recent research examined self-trapping in photorefractive crystals. Here, refractive index changes, which typically originate from the electro-optic effect, are saturable [8; 53]. This facilitates 1-D and 2-D self-trapping at relatively small intensities; entirely new forms of self-trapped light [8; 53] including dark [119; 120], spatially incoherent [121], and even white light solitons have also been discovered in photorefractive crystals [55].

The research presented in this Chapter examines the process of self-trapping in an organosiloxane medium, in which refractive index changes originate from a photo-initiated free-radical polymerisation reaction [24]. This photochemical approach provides opportunities to examine the dynamics and temporal evolution of self-trapped beams, which are entirely different from self-trapping dynamics in nonlinear optical (Kerr, photorefractive) materials. This is because of the following fundamental differences in the photoresponse between them (see Section 1.1.4): (i) index changes

in Kerr media decay immediately upon removal of the light field and in photorefractive crystals, at timescales determined by the dielectric relaxation time of the medium; polymerisation-induced refractive index changes are permanent and moreover, greater by at least an order of magnitude [24] (ii) polymerization can be initiated with a small amount of light-absorbing photoinitiator molecules; beam attenuation is therefore negligible. Depending on the wavelength and quantum yield of photoinitiation [19], self-trapping in photopolymers can in principle be achieved at a large range of wavelengths and at extremely low optical intensities (W/cm^2). Refractive index changes in Kerr media by contrast must be induced at intensities of GW/cm^2 . (iii) the photoresponse time in a polymerizable medium relies on photochemical reactions (ranging from ms to minutes) while in Kerr media, it is determined by a virtually instantaneous electronic response (10^{-15} s) and in photorefractive media, depend on the dielectric relaxation times, which can vary from ns to minutes.

Theoretical models based on the nonlinear Schrödinger equation have predicted several properties and trends in the behavior of self-trapped beams in polymerizable media [39; 20; 35]. These include the formation of multimode waveguides during self-trapping, the sequential excitation of high-order optical modes and the complex evolution of the modal composition of the self-induced waveguide as it evolves from single-moded to multimoded guidance. However, previous experimental studies of self-trapping in polymerizable media have been mainly concerned with channel waveguides induced by self-trapped beams, including tapered waveguides in epoxy resins [56] and cylindrical fibers in diacrylate and urethane-acrylate resins [57]. Related studies have also examined beam filamentation at increasing intensities [58; 57; 59], interactions of self-trapped beams [60; 57] and spectroscopic monitoring of waveguide-formation [61]. Potential applications of waveguides as single-mode [66], multimode [65] interconnects, fiber interconnects [23; 64] and components of wavelength division multiplexers [67; 68] have been examined. Complex self-written structures including replicas of high-order modes on fiber-tips [69], waveguide arrays [70] and even artificial compound eyes [72] have also been demonstrated. Collectively, the investigations listed above demonstrated that both 1-D [61] and 2-D [20; 60; 58] self-trapping could be achieved in polymerisation systems and highlighted potential applications of self-written structures. By contrast, the study presented here intends to systematically determine the temporal evolution of self-trapped beams and quantitatively characterise their dynamics over a broad range of intensities across 10 orders of magnitude (3.2×10^{-5} to $12732 \text{ W}/\text{cm}^2$), and compare these observations with existing theoretical predictions. Two significant features set this study apart: (i) while previous experiments characterised self-trapping through the structure and properties of self-induced waveguides or light scattered along the beam path, we employed beam profiling methods to directly visualise and quantitatively measure changes in the cross-sectional intensity profiles of the beam; (ii) the excellent spatial resolution in the photoresponse of the organosiloxane ($\sim 150 \text{ nm}$) [24] enabled quantitative experiments over a broad range of intensities without blurring, a diffusion-caused problem that has hampered previous studies based on liquid monomers [20; 58].

The Chapter is organized as follows. Detailed analysis of the observation of high order modes during self-trapping at a single intensity $1.6 \times 10^{-2} \text{ Wcm}^{-2}$ is first presented in Section 3.2. Trends elucidated from the intensity-dependence of self-trapping observed and spatial profiles at the entire range of intensities examined are then presented in Section 3.3. A summary of the diverse observed phenomena is presented in Section 3.4.

3.2 Self-trapping dynamics

3.2.1 Experimental verification

In a typical experiment, a linearly polarized, Gaussian laser beam at 532 nm with an average intensity of $1.6 \times 10^{-2} \text{ W/cm}^2$ was focused onto the entrance face of a transparent cuvette containing the photopolymerizable organosiloxane medium. Under linear conditions, the beam diffracted in both transverse directions from a focal width ($1/e^2$) of $20 \mu\text{m}$ to $118 \mu\text{m}$ (Figure 3.1a) as it propagated 6.00 mm from the entrance to the exit face of the medium. By contrast, in an organosiloxane sensitized to visible wavelengths with a titanocene photoinitiator ($\lambda_{max} = 393 \text{ nm}, 460 \text{ nm}$), the beam self-trapped and propagated without diffracting by initiating free-radical polymerisation of methacrylate groups and corresponding refractive index changes (Δn). The organosiloxane, which has a refractive index of 1.47, undergoes a maximum change in refractive index of $\Delta n_s \sim 0.006$ [24] (See also Chapter 2 for Experimental Methods).

Typical results are presented in Figures 3.1(b-r) and 3.2, which respectively are the 2-D spatial intensity profiles of the beam acquired at the exit face and the corresponding temporal plots of peak intensity and effective diameter. The beam self-trapped within 3 s, causing a 4.5-fold decrease in beam diameter from $118 \mu\text{m}$ to $31 \mu\text{m}$ and complementary increase in peak intensity from 0.133 mW/cm^2 to 0.46 mW/cm^2 . At 27 s, the beam narrowed further to $18 \mu\text{m}$ with an intensity of 1.30 mW/cm^2 ; the sharp contrast between the initially broad and diffracted beam and its subsequently self-trapped form is evident in Figure 3.1a and c. The self-trapped beam evolved over the next 388 seconds, exhibiting complementary oscillations in peak intensity and width (Figure 3.2). Corresponding spatial profiles showed that oscillations were due to the onset of high-order optical modes with characteristic spatial intensity profiles (Figure 3.1b-r). The modes observed during the evolution of the self-trapped beam were identified by comparison with the linearly polarized, high-order optical modes of passive cylindrical waveguides (Figure 3.3) which were computed using established models [122; 123]. Oscillations and individual modes were difficult to resolve (spatially and temporally) after 160 s. However, the beam remained self-trapped and did not revert to its diffracted form; at 415 s, its width was $23 \mu\text{m}$ (5-fold smaller

than the diffracted width). At this stage, there was an abrupt and irreversible decrease over the next 6 s in intensity to 0.4 mW/cm², which was probably caused by polymerisation induced phase separation in the organosiloxane.

3.2.2 Comparison with theoretical models

These experimental results do confirm predictions of theoretical models of self-trapping in polymerizable and equivalent one-photon based systems [20], [35], [124], [125], [63]. The theoretical framework summarized here is based on the nonlinear Schrödinger equation, which is generally applied to simulate nonlinear light propagation processes [117], [53]. The propagation of a Gaussian beam (with diameter $a \gg \lambda/2$, electric field amplitude E) according to the paraxial approximation,

$$E(x, y, 0, t) = E_0(-(x^2 + y^2)/a^2) \quad (3.1)$$

is given by

$$ik_0 n_0 \frac{\partial E}{\partial z} + \frac{1}{2} \nabla_t^2 E + k_0^2 n_0 \Delta n E + \frac{i}{2} k_0 n_0 \alpha E = 0 \quad (3.2)$$

where α is the attenuation coefficient of the medium at wavelength λ corresponding to the free space wavenumber, $k_0 = \omega/c$. The temporal variation of n due to polymerisation is given by a phenomenological expression [35],

$$\frac{\partial \Delta n}{\partial t} = A(EE^*) \left(1 - \frac{\Delta n}{\Delta n_s} \right) \quad (3.3)$$

where A is a material-dependent parameter and Δn_s , the maximum change of refractive index (at saturation) of the medium. Equation (3.3) reflects the kinetics of free-radical polymerisation, where the propagation rate is proportional to intensity (EE^*) and decays exponentially as the concentration of polymerizable monomers decreases (as $\Delta n/\Delta n_s \rightarrow 1$). This equation is equivalent to Equation (3.5) proposed by Kewitsch and coworkers. Equation (3.2) contains terms that describe the counterbalance between natural diffraction (x, y) and self-induced refractive index changes ($\Delta n(x, y, z, t)$) of the beam.

Numerical simulations of Equations (3.2) to (3.3) yielded temporal changes in the spatial distribution of refractive index and intensity along the propagation path (z) of the beam. Calculations of refractive index and intensity based on this same theoretical approach are presented in Figures 3.4 and 3.5 (See Chapter 6 for details on simulations).

From these results, the sequence and temporal evolution of the self-trapping process was proposed and trends in the modal evolution of the self-induced waveguide, identified. Simulations showed that self-trapping begins when the beam induces a gradient-index lens at the entrance face of the medium and self-focuses further along z as shown in steps 1 to 6 in Figure 3.4. From its new focal point, the beam induces

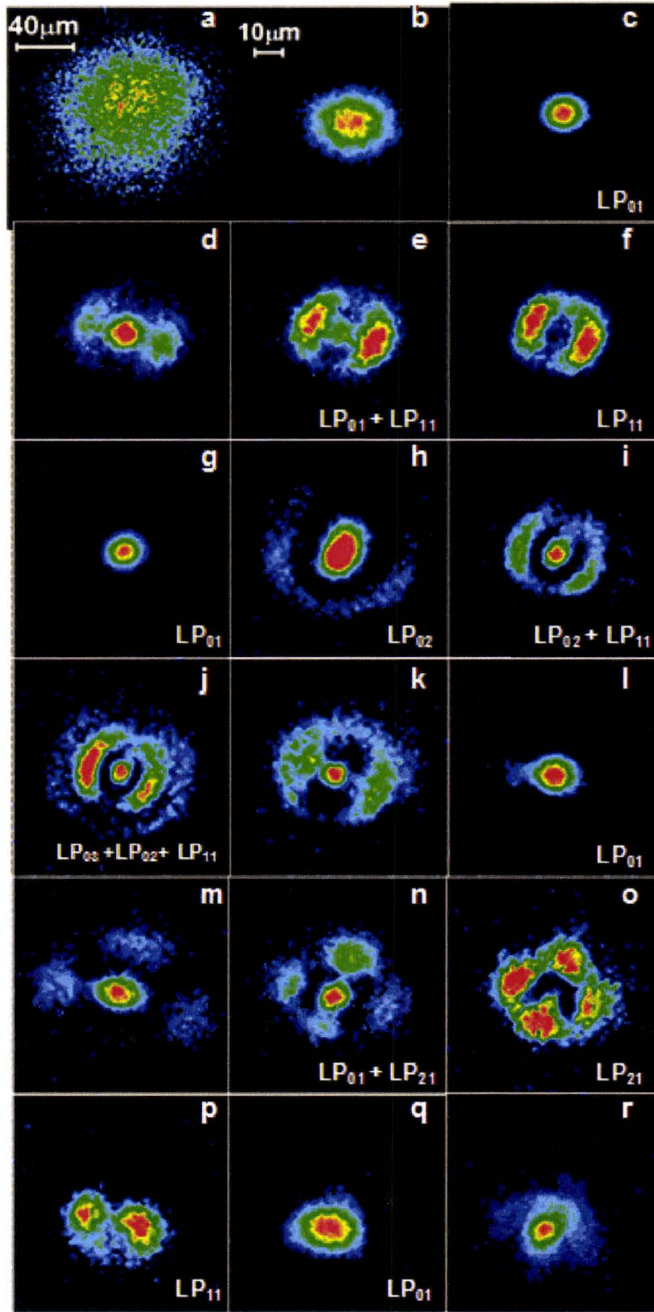


Figure 3.1: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00$ mm) during self-trapping in the organosiloxane at 1.6×10^{-2} W/cm². Profiles were acquired at (b) 3 s (c) 27 s (d) 30 s (e) 32 s (f) 35 s (g) 41 s (h) 47 s (i) 55 s (j) 57 s (k) 61 s (l) 116 s (m) 123 s (n) 129 s (o) 134 s (p) 141 s (q) 156 s and (r) 163 s. 2-D profile of the beam acquired under linear conditions (a) is included for comparison. Mode labels follow standard optical fiber nomenclature.

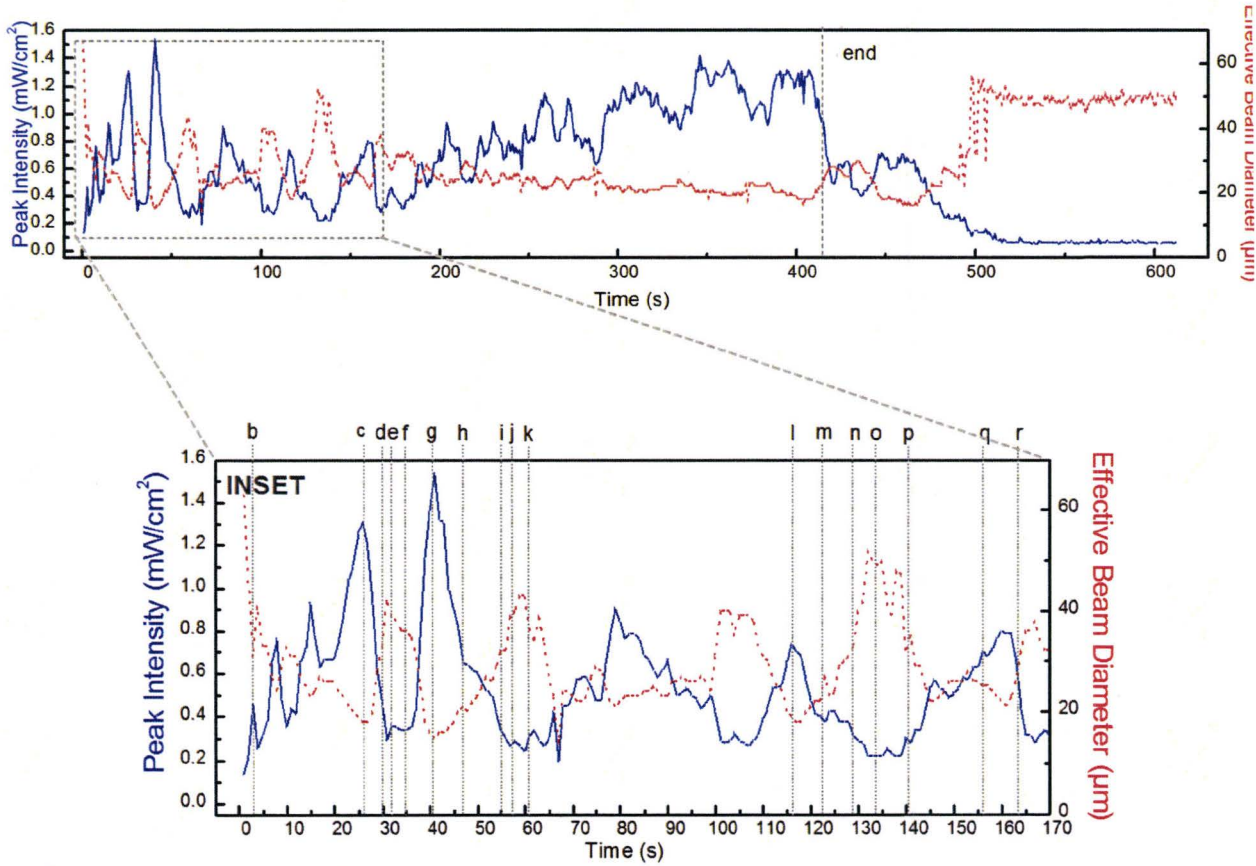


Figure 3.2: Temporal plots of peak intensity (solid-blue) and effective diameter (dotted-red) during self-trapping of a Gaussian visible (532 nm) laser beam at $1.6 \times 10^{-2} \text{ W/cm}^2$ in the organosiloxane medium. Termination of self-trapping is indicated by a vertical dotted line at 415 s. The inset contains the time-period (0 s - 170 s) corresponding to the onset of higher order modes. Times corresponding to 2-D spatial intensity profiles of the beam (Figure 3.1) are marked by dotted lines.

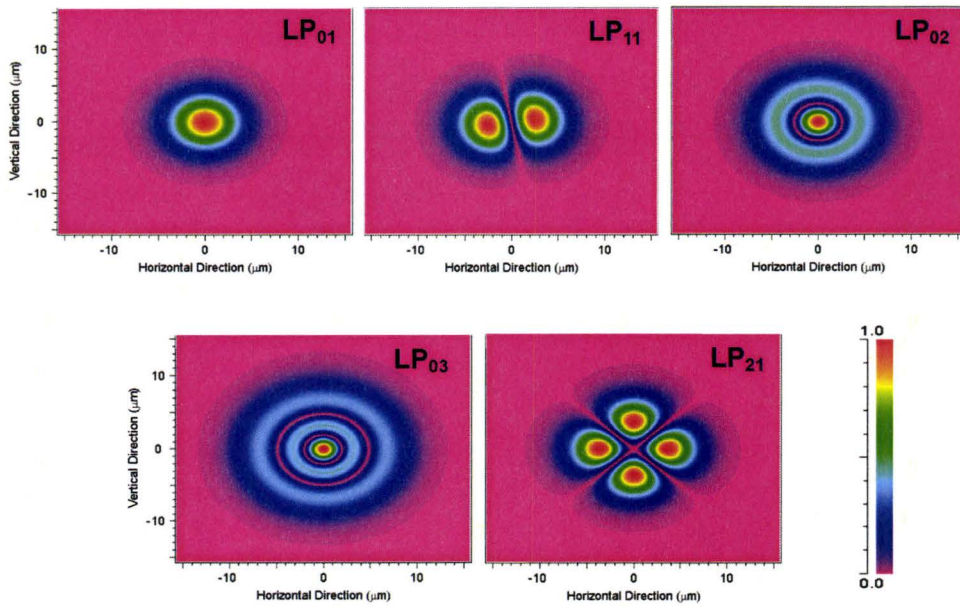


Figure 3.3: Beam propagation simulations of linearly polarized modes in an optical fiber performed with BeamPROPTM software. Modes were calculated for an optical fiber with diameter = $20\ \mu\text{m}$ and a Gaussian refractive index profile maximising at 1.479 at the axis and a cladding refractive index of 1.470. These values are comparable to the minimum and maximum values of refractive index changes in the photopolymerizable organosiloxane medium employed in studies of self-trapping.

Refractive index maps

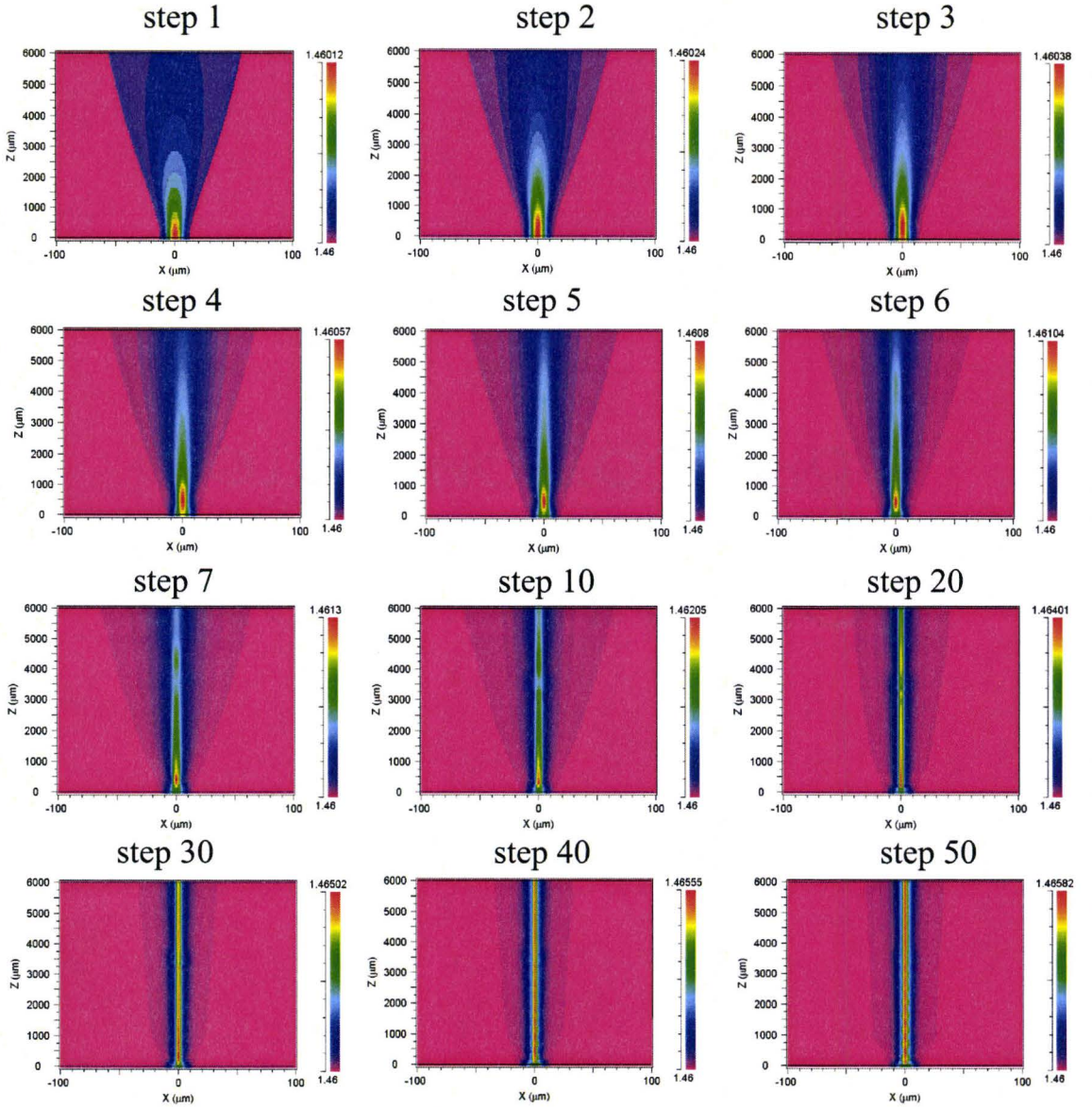


Figure 3.4: 2D Simulation results in Beamprop showing the refractive index profiles of a propagating Gaussian beam with $U_0 = 50$. The propagation coordinate is indicated with z and the transverse coordinate is x

Intensity maps

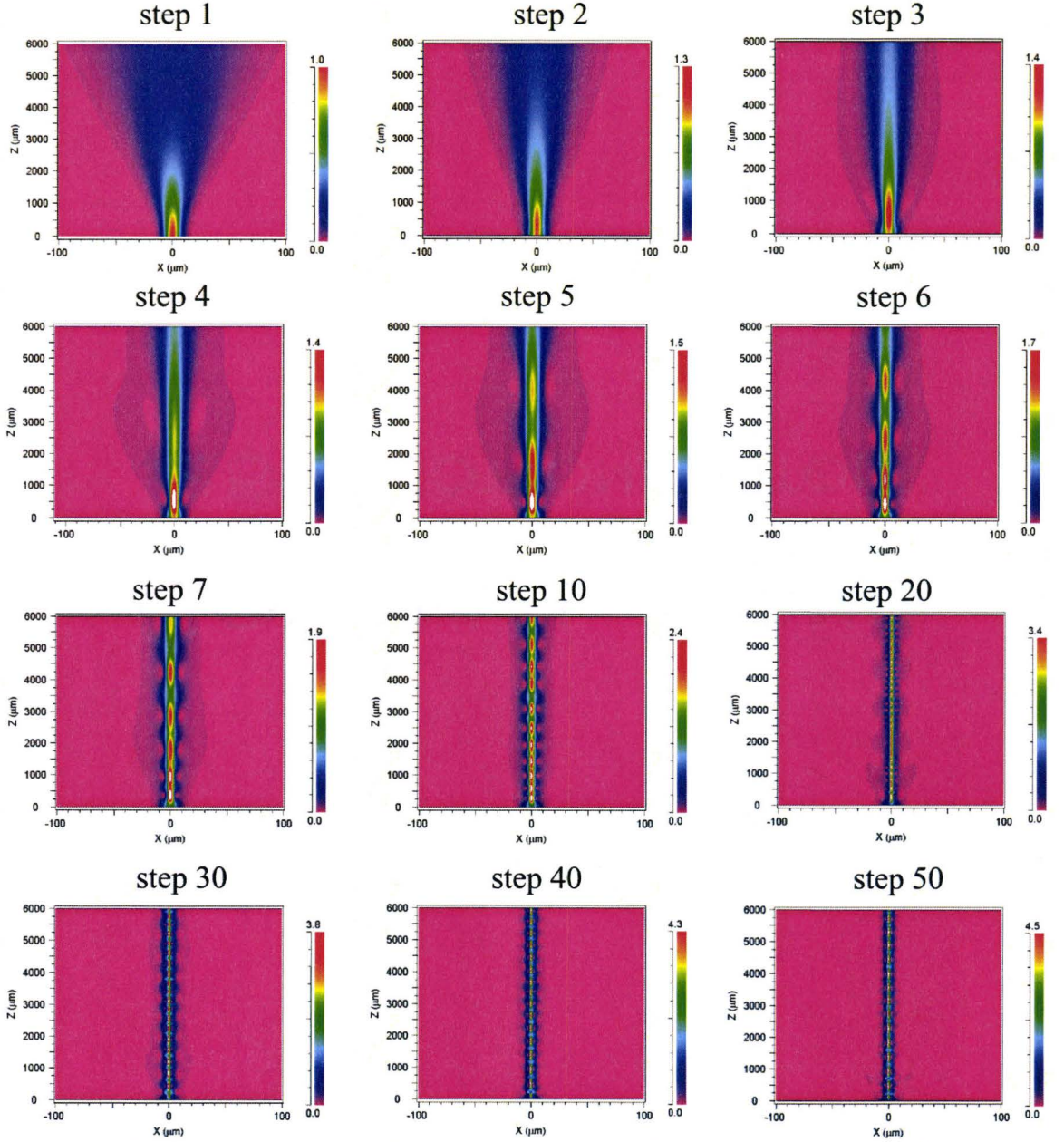


Figure 3.5: 2D Simulation results in Beamprop showing the intensity profiles of a propagating Gaussian beam with $U_0 = 50$. The propagation coordinate is indicated with z and the transverse coordinate is x

a gradient-index channel waveguide along its propagation path. The waveguide traps and guides the beam without diffracting to the exit face as observed in step 20 in Figure 3.5. Once self-trapped, the beam continues to increase the refractive index of its waveguide (steps 30 to 50 in Figure 3.4), which concomitantly develops multiple intensity maxima along z as observed in the intensity profiles of Figure 3.6. These maxima were attributed to the interference - beating - between high-order modes that become excited as the refractive index of the waveguide increases. Because refractive index changes are not uniform along z , the position and number of intensity maxima (and thus the modal composition) vary along the waveguide in a non-trivial way as can be seen in Figure 3.6, which shows the simulated transversal profiles at various points along z .

There is good agreement between the simulated sequence of self-trapping and the experimental observations presented in Figures 3.1 and 3.2. The decrease in width and increase in intensity observed within the first 30 seconds signifies self-focusing and subsequent self-trapping of the beam in its channel waveguide [20]. An optical micrograph acquired at this point confirmed that a cylindrical waveguide with a diameter of $\sim 40 \mu\text{m}$ had been inscribed along z (Figure 3.7a). Under linear conditions, the waveguide exhibited single-mode guidance (532 nm) with an output width ($1/e^2$) of $14.2 \mu\text{m}$ (Figure 3.7b).

Based on this, the average refractive index change (Δn) of the waveguide was approximated by using the condition for monomode operation of a waveguide with a gradient refractive index profile for the normalized frequency (V) [122; 126; 123]

$$V = \frac{2\pi a}{\lambda} \sqrt{n_1^2 - n_2^2} < 3.58 \quad (3.4)$$

where a is the radius of the waveguide, which in our case was taken as $20 \mu\text{m}$, based on Figure 3.7a. n_1 is the maximum refractive index at the core and n_2 the minimum refractive index at the cladding. The average refractive index change of the waveguide was calculated to be $< 7.8 \times 10^{-5}$, which is 2 orders of magnitude smaller than the refractive index change at saturation in the organosiloxane ($\Delta n_s \approx 0.006$) [24]. The self-trapped beam could therefore continue to raise the refractive index of its own waveguide over time, which in turn led to the sequential excitation of high-order optical modes. That the self-trapped beam evolves in this way is in itself different from the behavior of 2-D self-trapped beams in photorefractive media. Δn_s in photorefractive crystals is small ($\approx 10^{-4}$) [117] and the system generally saturates upon self-trapping; because no further change in refractive index is possible, the self-trapped beam remains stable in intensity and width.

During self-trapping in the organosiloxane, three different high-order modes appeared in sequence and were individually observed at the exit face (Figure 3.1). Briefly, at 27 s, the fundamental LP_{01} mode (Figure 3.1c) was succeeded at 35 s by the next-order mode LP_{11} (Figure 3.1f) followed by the LP_{02} and LP_{21} modes, which appeared at 47 s and 134 s, respectively (Figures 3.1h, and 3.1o). Superpo-

Profiles of step 50 at various "z"

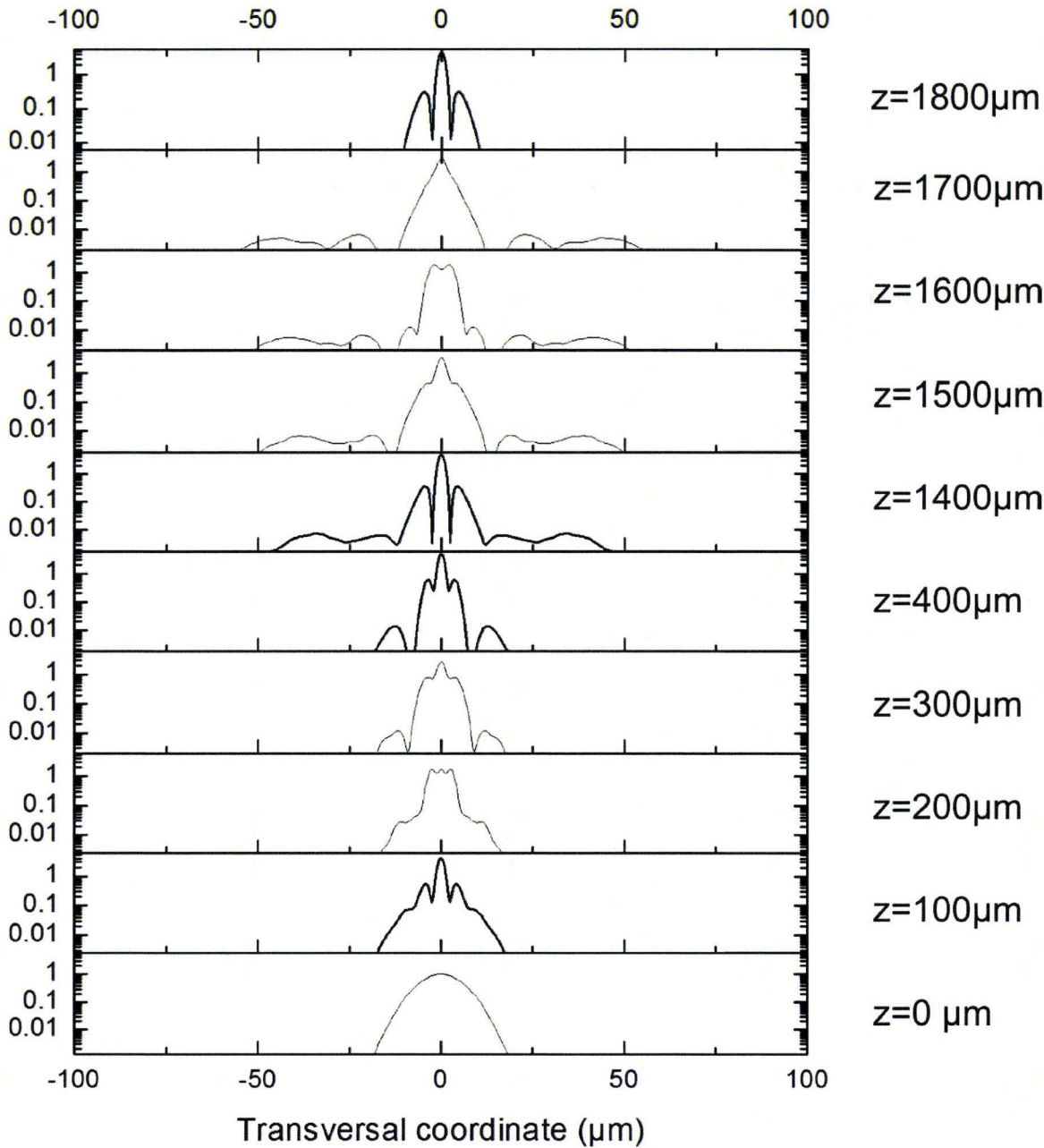


Figure 3.6: Simulated intensity profiles at different z values along the propagation axis corresponding to initial beam and two oscillations for step 50 of a propagating Gaussian beam with $U_0 = 50$. The relative intensity is shown on a logarithmic scale for clarity. Intensity peaks are shown in bold.

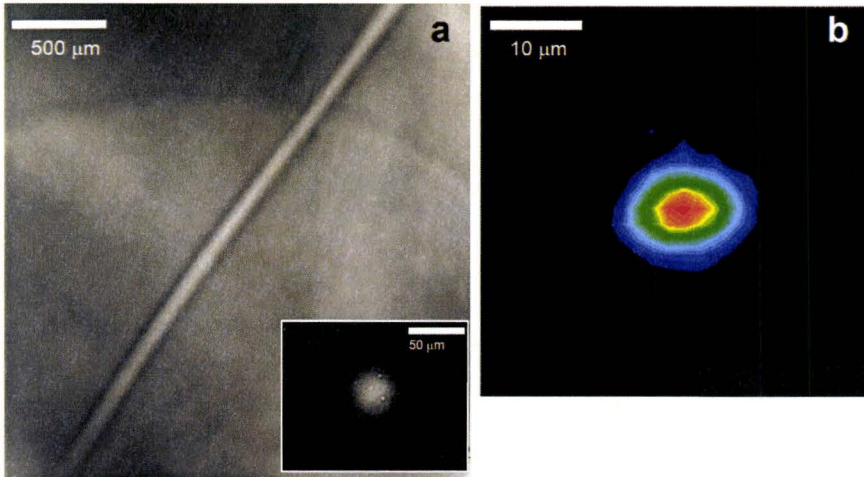


Figure 3.7: (a) Transmission optical micrograph of the self-written waveguide in the organosiloxane during self-trapping at $1.6 \times 10^{-2} \text{ W/cm}^2$ (average power 50 nW); the inset is a micrograph of its transverse (x, y) cross-section. (b) 2-D intensity profile at the output ($z = 6.0 \text{ mm}$) of the waveguide guiding light at 532 nm under passive conditions; the output is Gaussian with diameter $(1/e^2) = 14.2 \text{ μm}$. Micrographs were acquired through a red filter to prevent polymerization during measurement.

sitions of high-order modes, including a fourth high-order mode LP_{03} with the LP_{01} mode were also observed (Figures 3.1e, 3.1i, 3.1j, 3.1m, 3.1n). Comparisons with computed profiles shown in Figure 3.3 demonstrated that they corresponded to the linearly-polarised modes characteristic of cylindrical waveguides such as optical fibers [122; 126; 123]. This is consistent with the cylindrical waveguides induced by self-trapped beams (Figures 3.7 and 3.9). In numerical simulations, the excitation of high-order modes was also confirmed through intensity profiles along z as seen in Figure 3.6 and by plotting the intensity profiles at the output of the medium over time as shown in Figure 3.8. In these simulations, the profile along z does not remain Gaussian, instead the beam presents multiple lobes which can be attributed to superposition of optical modes. Unlike previous simulations where multiple modes were inferred only from the emergence of intensity maxima along the propagation axis (z) of the waveguide and corresponding calculations of modal propagation constants [63], our simulations also revealed the transversal intensity profile. Results from current experiments not only confirm the onset of high-order modes but also enable direct visualization of their spatial intensity profiles in the near-field and *in situ* monitoring of their evolution within the waveguide.

Numerical simulations (Figures 3.4, 3.5, 3.6 and 3.8) based on Equations (3.1) to (3.3), show that multiple high-order modes propagate within the same self-written channel waveguide. The simulations also show that the spatial variations in refractive index occur at smaller rates and are consequently less pronounced than corresponding variations in optical intensity (Figures 3.4 and 3.5). The self-induced waveguide

can therefore host high-order modes with non-Gaussian intensity profiles, even while retaining its cylindrical geometry. In fact, the same property has enabled self-trapping of incoherent white light, which is composed of a randomly and rapidly fluctuating distribution of optical modes, to collectively propagate within a single self-induced waveguide in the organosiloxane [27; 127].

To further confirm experimentally that the multiple modes were all excited within the same cylindrical waveguide, optical micrographs were acquired after excitation of LP_{11} and separately, at longer times after excitation of LP_{21} (Figures 3.9b and 3.9e). Both revealed a single cylindrical waveguide: the waveguide formed after excitation of LP_{11} had an overall diameter of $72\ \mu\text{m}$ while that formed after the onset of LP_{21} was 10-fold wider with an overall diameter of $240\ \mu\text{m}$. Both guided light (532 nm) under passive conditions; guidance was strongly confined to the core with effective output-diameters of $32\ \mu\text{m}$ and $30\ \mu\text{m}$, respectively (Figures 3.9c and 3.9f). This is consistent with theoretical simulations, which showed that self-induced waveguides possess a gradient refractive index profile and are thus able to efficiently confine light within their core-regions (Figure 3.4).

3.2.3 Oscillations of the self-trapped beam

Numerical simulations of Equations (3.1) to (3.3) have predicted that a self-trapped beam in a polymerizable medium would always exhibit oscillatory behavior, due to the evolving modal structure of the self-induced waveguide and the consequent changes in its intensity distribution. (The same changes can be followed in simulations presented in Figures 3.5 and 3.8). Such theoretical predictions are understandable because interference between two even modes in a channel waveguide generates intensity maxima positioned at intervals along z with a periodicity of $2\pi/|\beta_1 - \beta_2|$, where β_1 and β_2 are the respective propagation constants of the modes [63]. Such maxima would be periodically positioned in a waveguide with a uniform refractive index profile. The refractive index of a self-induced waveguide however varies significantly along z ; as a result, both the number of modes and the propagation constant of each individual mode vary along z , which is further confirmed through Wentzel-Kramers-Brillouin analysis [63]. Intensity maxima due to mode-beating are therefore positioned aperiodically along z . In a passive waveguide, such maxima would remain stationary. However, due to the continually changing refractive index profile of the self-induced waveguide, these maxima change position, appearing to translate along z over time. At a constant observation point along z , the apparent translation of maxima would lead to aperiodic oscillations of the overall intensity and width of the self-trapped beam.

Such oscillations of intensity and width were indeed observed in the current study, experimentally and numerically, where the self-trapped beam was monitored over time at a constant point along z at its exit face (Figure 3.10 and 3.8). These results gave further insight into the modal evolution of the self-induced waveguide that were not

Profiles at $z=6.00\text{mm}$

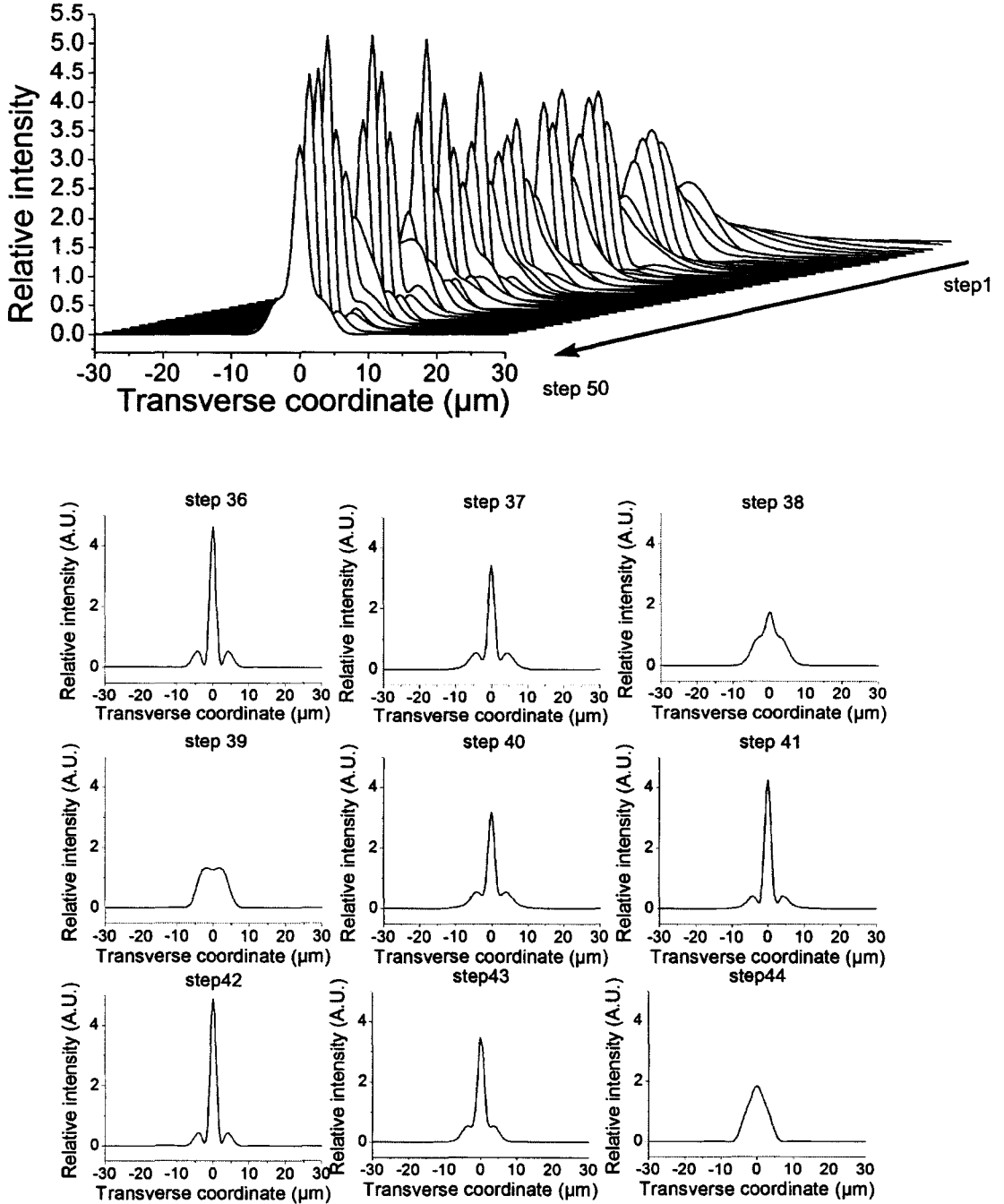


Figure 3.8: Profiles of intensity for $U_0 = 50$ in the transversal direction x at the output of the medium ($z = 6.00\text{mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step in the computation represents a relative unit time.

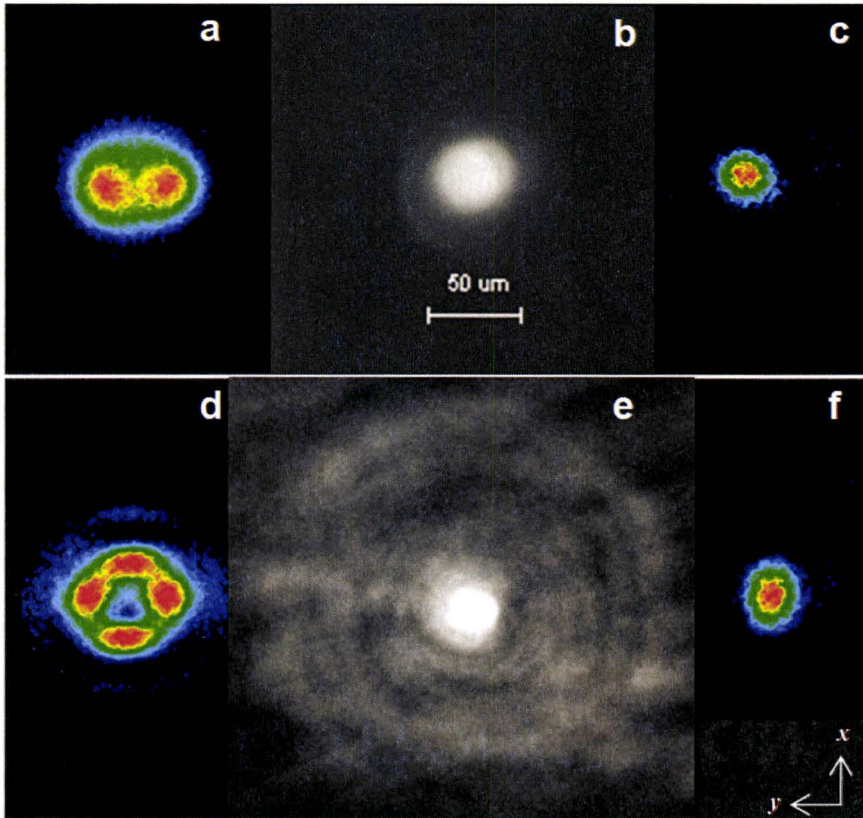


Figure 3.9: Characterisation of self-written waveguides after the onset of higher order modes during self-trapping at $1.0 \times 10^{-2} \text{ W/cm}^2$. (b) Transmission optical micrograph of self-written cylindrical channel waveguide after the onset of (a) LP_{11} . (c) Intensity profile of waveguide output ($z = 6.0 \text{ mm}$) under passive conditions; output diameter ($1/e^2$) = $37.8 \mu\text{m}$. (e) Cross-sectional micrograph of self-written waveguide after onset of (d) LP_{21} . (f) Intensity profile of waveguide output ($z = 6.0 \text{ mm}$) under passive conditions; output diameter ($1/e^2$) = $36.0 \mu\text{m}$. Micrographs were acquired through a red filter to prevent polymerization during measurement. The scale bar in (b) applies to all images.

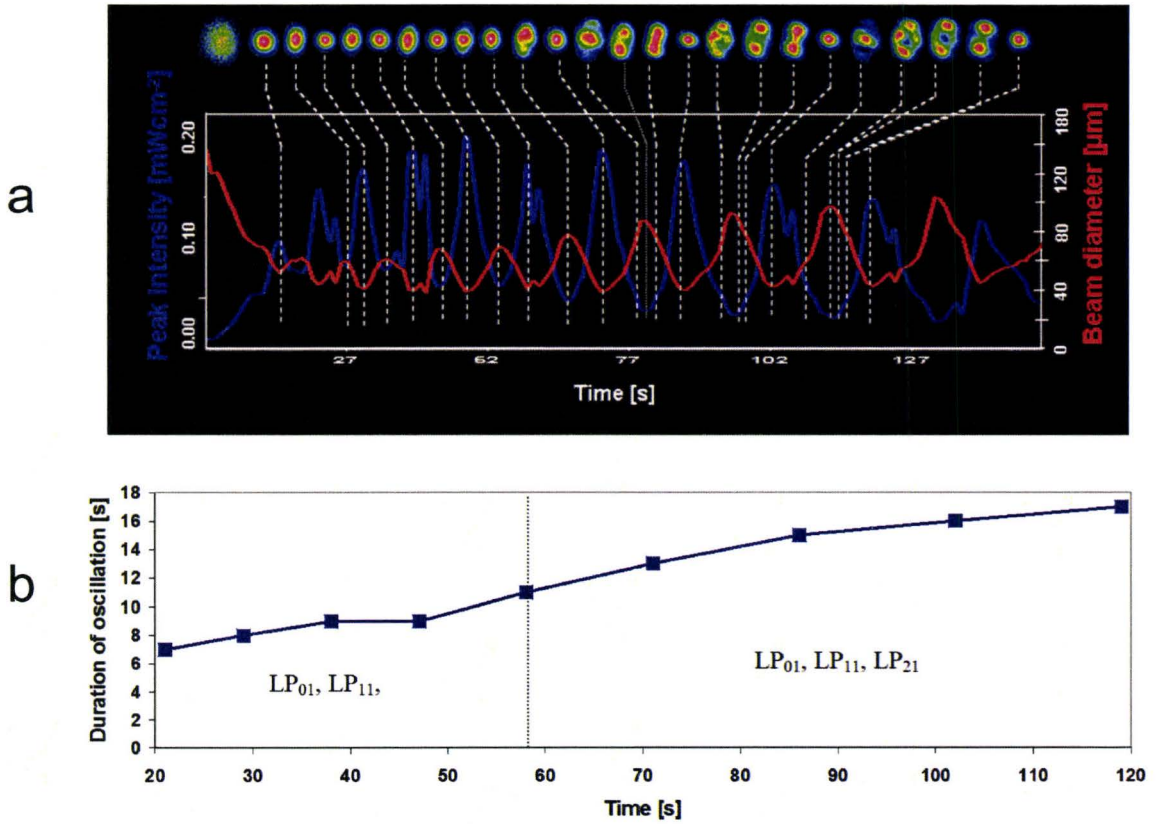


Figure 3.10: (a) Temporal changes of beam width and peak intensity and corresponding evolution of modal composition at $z = 6.0$ mm during self-trapping at 3.2×10^{-3} W/cm². (b) Duration of oscillations over time. The modes identified within each set of oscillations are indicated

evident in previous theoretical simulations. For example, previous theoretical models considered the excitation of only one high-order mode [63].

Furthermore, the excitation of the second mode was inferred by its interference with the first and consequent intensity variations along z ; the spatial intensity profiles corresponding to individual modes were not obtained. By contrast, the experimental results obtained in this study enabled the direct visualisation of each of the high-order modes, identifying up to five discrete modes and moreover confirming that they corresponded to optical fibre modes (Figure 3.1). It was therefore possible, as described below, to identify the exact sequence of optical modes that corresponded to each oscillation of the beam and in this way, observe the evolution of the modal composition at the waveguide at its output. To our knowledge, there are no previous examples of direct monitoring of the evolution of a cylindrical waveguide from single-mode to multimode guidance.

The oscillatory dynamics of the self-trapped beam varied with input intensity. Because the rate of refractive index change decreased with average intensity, there

was a corresponding decrease in both the number of modes and the rate at which the modal composition of the self-written waveguide changed (see Figures 3.15 and Figures 3.16 to 3.19). While oscillations of the self-trapped beam at $1.6 \times 10^{-2} \text{ W/cm}^2$ were rapid and irregular (Figure 3.2), they could be better resolved by carrying out self-trapping at a lower intensity, $3.2 \times 10^{-3} \text{ W/cm}^2$, where the rate of refractive index changes (and thus rate of self-trapping) is significantly lower.

As observed in Figure 3.10a, the crest of an oscillation corresponded to the brightest and narrowest mode, LP_{01} , and the valley, to the highest mode in the sequence. Superposed modes fell between the valleys and crests. Briefly, the first 5 oscillations showed alternation between modes LP_{01} and LP_{11} ; once LP_{21} was excited at 67 s; the next 4 oscillations each consisted of the sequence: LP_{01} , LP_{21} , LP_{11} , LP_{01} . Transitions between the pure modes were superpositions of the two: for e.g., superposition $LP_{11} + LP_{01}$ appeared at 83 s during the transition from LP_{01} to LP_{01} while $LP_{21} + LP_{11}$ appeared at 115 s between LP_{21} and LP_{11} . Numerical simulations shown in Figure 3.8 corresponding to intensity profiles at the output of the medium also show an oscillatory behavior. High-order modes are present most of the time as seen in the individual profiles from steps 36 to 44. However, when the overall intensity is small, only a single high-order mode is present as seen in steps 38 and 39. When the overall intensity is large either a superposition of modes or LP_{02} is observed as seen in steps 36 and 42, here side lobes are much smaller in intensity. This could be the reason why in experimental observations only the fundamental mode is observed (Figures 3.1c, g, l and q). A plot of the experimental duration of each oscillation against time showed the theoretically predicted aperiodicity of oscillations and its general increase over time (Figure 3.10b). The latter can be attributed to the decrease in the rate of refractive index changes, which decreases the rate of change of modal composition and thus the oscillations of the self-trapped beam. Although the sequence of excitation of high-order modes over time corresponds to the sequence supported by optical fibers with increasing indices of refraction, their specific order of appearance, recurrence and superpositions cannot be fully explained by the existing theoretical models. Further theoretical modeling, which takes into account changes to the intensity distribution in 3-D is necessary to fully rationalize the specific sequence of appearance of high-order modes.

3.3 Intensity dependence

3.3.1 Importance of photoinduced refractive index profile

The previous Sections described self-trapping of the beam with an incident intensity of $1.6 \times 10^{-2} \text{ W/cm}^2$. We then examined nonlinear laser propagation in the organosiloxane at three intensity regimes that spanned 10 orders of magnitude (Fig-

Average Intensity [W·cm ⁻²]	Average power [W]	
3.2x10 ⁻⁵	1x10 ⁻¹⁰	Low intensity regime
1.6x10 ⁻⁴	5x10 ⁻¹⁰	
0.003	10x10 ⁻⁹	
0.008	25x10 ⁻⁹	
0.016	50x10 ⁻⁹	
0.19	600x10 ⁻⁹	Mid-intensity regime
1.6	5x10 ⁻⁶	
16	50x10 ⁻⁶	
159	500x10 ⁻⁶	High intensity regime
1592	5x10 ⁻³	
12732	40x10 ⁻³	

Figure 3.11: Range of intensities (left column) and corresponding average powers (right column) for self-trapping experiments covering 10 orders of magnitude in intensity.

Figure 3.11): low (3.2×10^{-5} , 1.6×10^{-4} , 3.2×10^{-3} , 8.0×10^{-3} , 1.6×10^{-2} W/cm²), mid (0.19, 1.6, 16 W/cm²) and high (159, 1592, 12732 W/cm²); the beam was Gaussian and linearly polarized with a width ($1/e^2$) of 20 μ m at all intensities.

Results showed that the dynamics of self-trapping were strongly dependent on intensity. Significantly, they also revealed the existence of other nonlinear phenomena including diffraction rings, single ring formation and filamentation at the mid and high intensity regimes.

The importance of the refractive index profile originates from the expression for polymerization-induced refractive index change [20],

$$\Delta n(x, y, z, t) = \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(t)|^2 dt \right] \right\} \quad (3.5)$$

where Δn_s is the maximum refractive index change, U_0 is the critical exposure required to initiate polymerization, τ is the monomer radical lifetime and $|E(t)|^2$ is the square of the electric field amplitude or intensity (I) of the incident optical field and t is time.

By plotting Equation (3.5) (Figure 3.12a), we observe that the refractive index change increases with light exposure, until saturation, where it maximises (Δn_s). According to Equation (3.5), Δn_s can be achieved with high and low intensity beams at relatively short and long times, respectively.

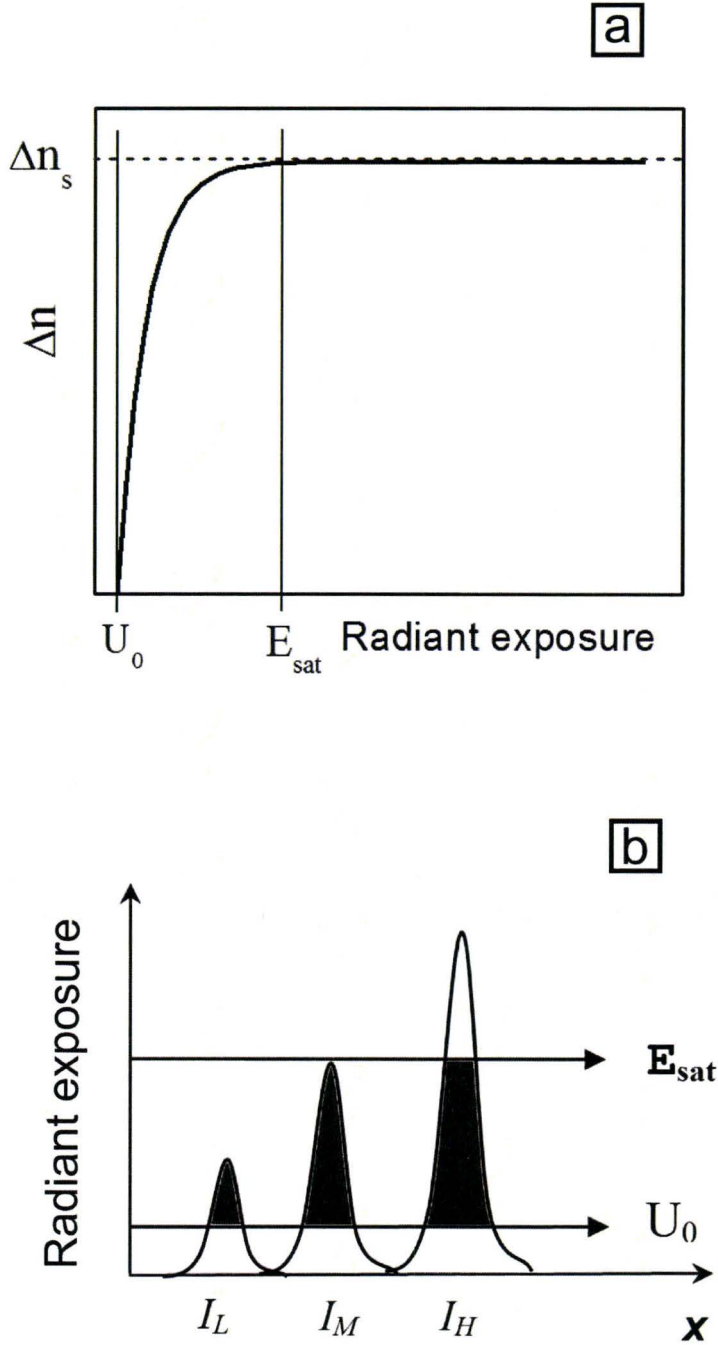


Figure 3.12: Plots of Equation (3.5) (a) Refractive index change as a function of radiant exposure, where U_0 = critical exposure required for photoinitiation and E_{sat} = radiant exposure required to achieve Δn_s = refractive index change at saturation. (b) 1-D spatial profiles (along x) of Gaussian beams and corresponding optical radiant exposure after one second of irradiation, for low (I_L), medium (I_M) and high (I_H) intensity beams. Relative positions of E_{sat} and U_0 are indicated. Induced refractive index profiles are filled in dark grey.

Schemes of refractive index profiles induced by Gaussian beams of low, mid and high intensities are presented in Figure 3.12b. After irradiation for the same period of time, the index induced by the low intensity beam (I_L) remains Gaussian maximising at a value $< \Delta n_s$ below saturation point. At mid-intensity (I_M) the index profile remains Gaussian although its maximum is closer to Δn_s . At high intensity (I_H), the index profile is no longer Gaussian. Here the intensity of the beam is sufficient to induce Δn_s over a large area of the beam (and not only at its axis). This leads to a top-hat or flattened Gaussian profile of the refractive index.

3.3.2 Quantitative trends

Results at each intensity are presented as temporal plots of beam diameter and peak intensity monitored at the exit face of the medium; most significant changes in the corresponding 2-D spatial intensity profiles are presented in Section 3.3.3. To quantitatively compare self-trapping dynamics in the different intensity regimes, temporal plots were analysed in terms of 3 selected parameters (a) self-focusing time, (b) self-trapped beam width and (c) transmittance ($I_{z=6.0mm}/I_{z=0.0mm} * 100\%$). Each value was averaged over at least three repeat experiments at each intensity; corresponding values of standard deviation are provided (Figures 3.13 and 3.14).

3.3.2.1 Self-focusing time

The self-focusing time was defined as the time taken by the diffracted beam (at $t = 0$ s) to first exhibit significant narrowing. In each temporal plot of self-trapping, this corresponded to the initial steep decrease in width and complementary increase in peak intensity as can be seen in Figure 3.2.

The plot of self-focusing time against initial intensity is approximately parabolic (Figure 3.13a). In the low intensity regime, self-focusing time varied inversely with intensity, decreasing from 200 ± 90 s to 11 ± 3 s as the initial intensity of the beam was increased from 3.2×10^{-5} W/cm² to 8.0×10^{-3} W/cm². From this minimum, it increased to 21 ± 9 s to 140 ± 30 s in the mid-intensity regime and more significantly to 1300 ± 500 s at 1592 W/cm² before decreasing to 300 ± 200 s at the greatest intensity of 12732 W/cm².

To self-focus, the beam must induce refractive index changes in the form of its own Gaussian profile at the entrance face of the medium. Figure 3.12b depicts how such lenses are induced at intensities in the low, mid and high-intensity regimes. The strongest lens would possess the steepest gradient, with the maximum possible refractive index (Δn_s) localized to the axial region with a radially symmetric decay from this point. In the low-intensity regime, long times ($t \gg \tau$) are necessary to achieve Δn_s even at the (most intense) axial region of the beam. Because I and t vary inversely (until Δn_s is reached), the time required to induce a lens with the

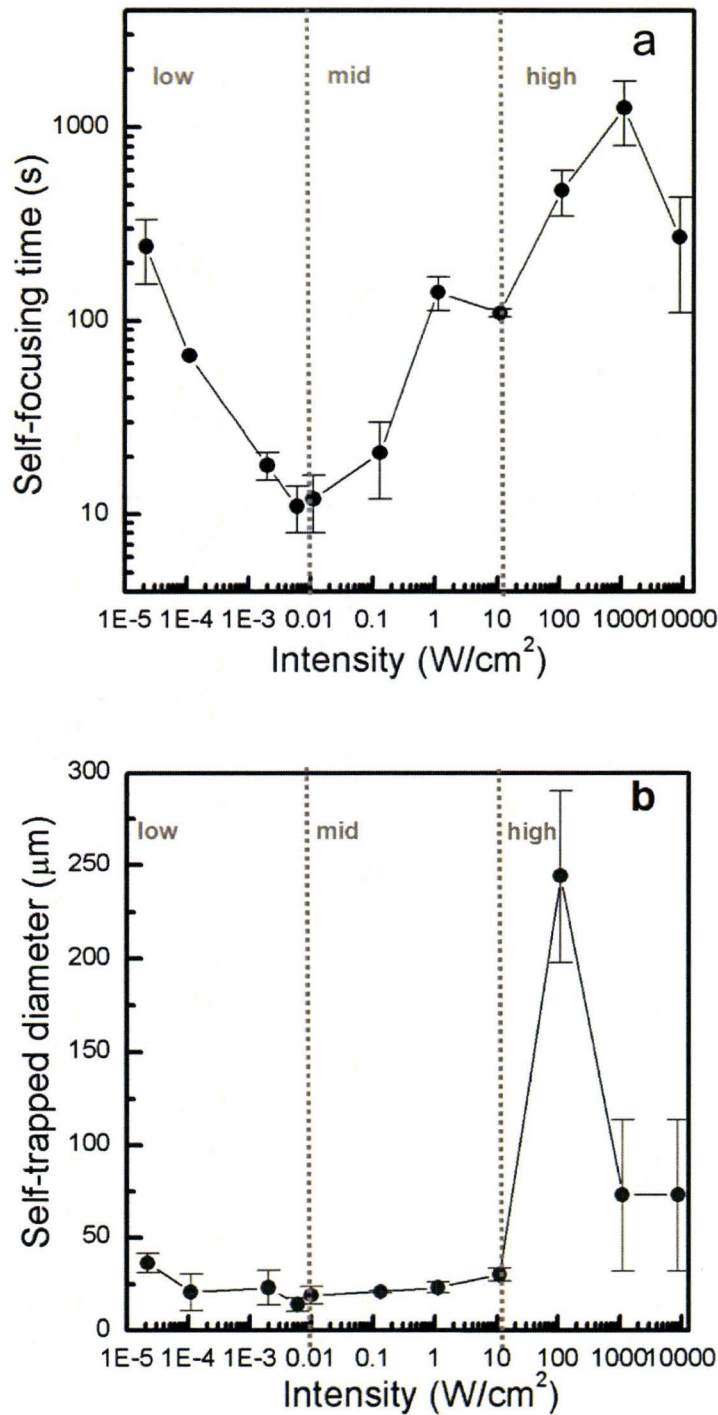


Figure 3.13: Plots of the (a) self-focusing time and (b) self-trapped diameter versus the initial average intensity of the beam. The low, mid and high-intensity regimes are marked with dotted lines. Error bar = $1 \times \sigma$.

strongest gradient also varies inversely with intensity within the low-intensity regime, leading in turn to the inverse relationship between self-focusing time and intensity (Figure 3.13a). The trend is reversed in the mid and high-intensity regimes, where increasingly larger cross-sections of the beam simultaneously achieve saturation; Δn_s is therefore delocalized over a larger area. Lenses are wider and weaker with smaller numerical apertures that collect and focus light less efficiently ; self-focusing consequently requires increasingly longer times.

3.3.2.2 Self-trapped diameter

The plot of self-trapped beam width (Figure 3.13b) follows the same trend as the intensity-dependence of self-focusing time (Figure 3.13a) and is consistent with the mechanism proposed in Figure 3.12. The self-trapped beam width decreased from $31 \pm 4 \mu m$ to a minimum of $12 \pm 3 \mu m$ as the intensity was increased from $3.2 \times 10^{-5} \text{ W/cm}^2$ to $8.0 \times 10^{-3} \text{ W/cm}^2$; this corresponded to a 4.0 ± 0.5 to 11 ± 3 -fold decrease in diameter relative to the diffracted beam. In the low intensity regime, lenses with steeper index gradients and thus larger numerical apertures are induced at increasing average intensity leading to more efficient suppression of diffraction and better confinement of light. As observed in Figure 3.13b, this trend was reversed in the mid and high-intensity regimes. Here, the index gradient weakens with increasing intensity; light diffraction is less efficiently suppressed leading in turn to an increase in the self-trapped beam width. Accordingly, in the mid-intensity regime (0.19 W/cm^2 to 16.00 W/cm^2), there was a general increase in the self-trapped beam width, which ranged from $18.0 \pm 0.6 \mu m$ to $25 \pm 3 \mu m$. In the high intensity regime, the self-trapped beam width was $70 \pm 40 \mu m$ at 1592 W/cm^2 and effectively no decrease in width was observed at the greatest intensity of 12732 W/cm^2 (The anomalous increase in self-trapped beam width to $200 \pm 50 \mu m$ at 159.0 W/cm^2 is associated with the beam filamentation as detailed in Section 3.3.3.4).

3.3.2.3 Transmittance

The efficiency of light confinement and guidance of a passive channel waveguide depends on the magnitude and spatial profile of its core refractive index; transmittance is greater in a waveguide with a gradient index relative to a uniform (step) index profiles [123]. The greater rate of self-focusing and smaller self-trapped widths observed in the low intensity regime (Figure 3.13) were attributed to steeper index gradients. It follows that the absolute amount of light that is transmitted through, the waveguide ($I_{z=6.0mm}/I_{z=0.0mm} * 100\%$, the percentage of intensity transmitted at $I_{z=6.0mm}$) should also be enhanced during self-trapping in the low intensity regime. This was confirmed in Figure 3.14, which is a plot of transmittance versus initial intensity of the beam. There was an initial increase in transmittance in the low in-

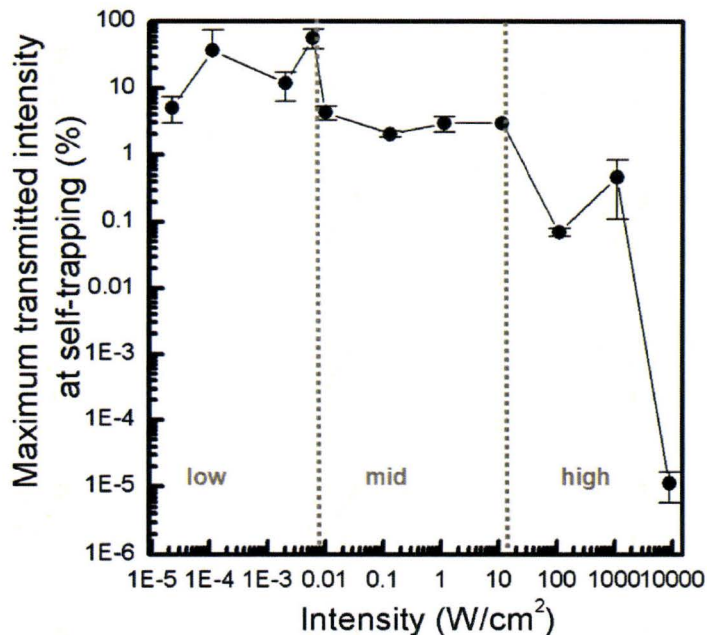


Figure 3.14: Plot of the maximum transmitted intensity at self-trapping versus initial average intensity of the beam. The low, mid and high-intensity regimes are marked with dotted lines. Error bar = $1 \times \sigma$.

tensity regime followed by an overall decrease. The maximum transmittance of $56 \pm 18 \%$ occurred during self-trapping at 0.008 W/cm^2 ; which was the intensity at which the most rapid self-focusing and smallest self-trapped diameter were observed.

3.3.2.4 Reproducibility

In the discussion above, each point constituting plots in Figures 3.13 and 3.14 was an average taken from at least thrice-repeated experiments. It is important to note that the self-trapping process in each repeat experiment was identical (that is, spatial and temporal variations of the beam followed identical trends). When quantitatively analysed, these trends were remarkably reproducible at the low and mid-intensity regimes, particularly in terms of self-trapped beam width. However, the standard deviation was in general large for measurements made in the high intensity regime. Free-radical polymerization is an exothermic process, which due to increased rates at high intensities may lead to convection currents and inhomogeneities in the medium that contribute to variations between experiments, which were conducted under ambient conditions. However, it is equally important to note that detailed quantitative analyses of self-trapping have not previously been carried out. The results presented here both provide insight into the mechanism of self-trapping and also a quantitative evaluation of its reproducibility

3.3.3 Spatial beam profiles: high order modes, spatial diffraction rings, single ring formation and filamentation

As shown in Section 3.3.1, the refractive index profile induced in the organosiloxane varies with input intensity as a result of the Δn saturation. This ultimately leads to the observation of different phenomena.

2-D spatial intensity profiles acquired during self-trapping at different intensities showed striking differences in the behavior of beams within each intensity regime. 2-D profiles are shown in the following Sections; representative profiles are presented in Figure 3.28.

3.3.3.1 Low intensity regime: high order modes

In the low-intensity regime, self-trapping at all intensities followed the theoretically predicted sequence of self-focusing, waveguide formation and excitation of high-order modes (Figures 3.16, 3.17, 3.18, 3.19), this last one accompanied with oscillations of the beam (Figure 3.15). The principal difference was the absolute number of high-order modes excited in the waveguide. Because the rate of refractive index change decreases with decreasing intensity (Equation (3.5)), waveguides induced at smaller intensities have smaller values of refractive index change and can therefore support fewer modes [123]. For example, only three modes (LP_{01} , LP_{11} and LP_{21}) were identified at the three lowest intensities (3.2×10^{-5} , 1.6×10^{-4} and 3.2×10^{-3} W/cm²) whereas five modes (LP_{01} , LP_{11} , LP_{21} , LP_{02} and LP_{03}) were evident during self-trapping at 8.0×10^{-3} and 1.6×10^{-2} W/cm².

3.3.3.2 Mid-intensity regime: diffraction rings

The resolution of high-order modes was not possible during self-trapping in the mid-intensity regime. Here, refractive index changes occurred at greater rates and the entire self-trapping process was completed within a significantly reduced period of time. (For example, the average duration of self-trapping * was 703 s at the low intensity of 1.6×10^{-2} W/cm² whereas it was only 184 s at the mid intensity of 16 W/cm²). As a result, the self-induced waveguide rapidly achieves saturation and is rendered multimoded at early times, as observed at the output during self-trapping at 0.19 W/cm² (Figure 3.20e).

At very long times (≈ 500 s), there was a re-emergence of a single peak (Figure 3.20i) that sustained its profile until 605 s; in the corresponding temporal plot (Figure 3.21a), this was indicated by an increase in peak intensity and corresponding decrease beam width at 486 s. The new peak was positioned at a distance of $163 \mu\text{m}$, 45° to

*The duration of self-trapping was defined as the time over which the beam remained significantly narrower (at least $>\approx 1.5$ fold) and more intense than its diffracted form.

Low intensity regime

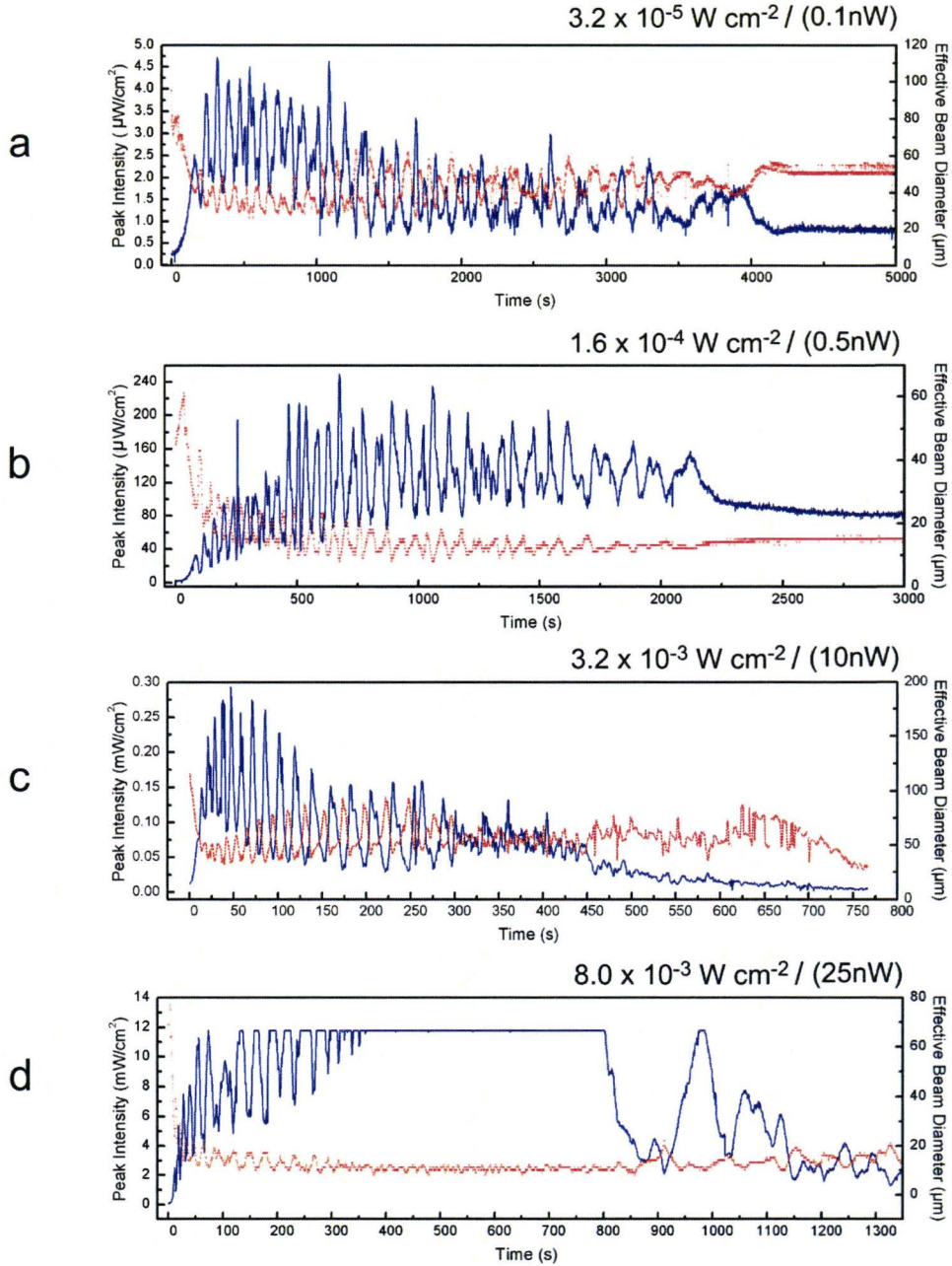


Figure 3.15: Graphs of temporal evolution of peak intensity (solid blue line) and effective beam diameter, $1/e^2$ (dotted red line) collected at the exit face of the organosiloxane corresponding to the low intensity regime: a) $3.2 \times 10^{-5} \text{ W/cm}^2$ (power 0.1 nW), b) $1.6 \times 10^{-4} \text{ W/cm}^2$ (power 0.5 nW), c) $3.2 \times 10^{-3} \text{ W/cm}^2$ (power 10 nW) and d) $8.0 \times 10^{-3} \text{ W/cm}^2$ (power 25 nW).

$$3.2 \times 10^{-5} \text{ W cm}^{-2} / (0.1 \text{ nW})$$

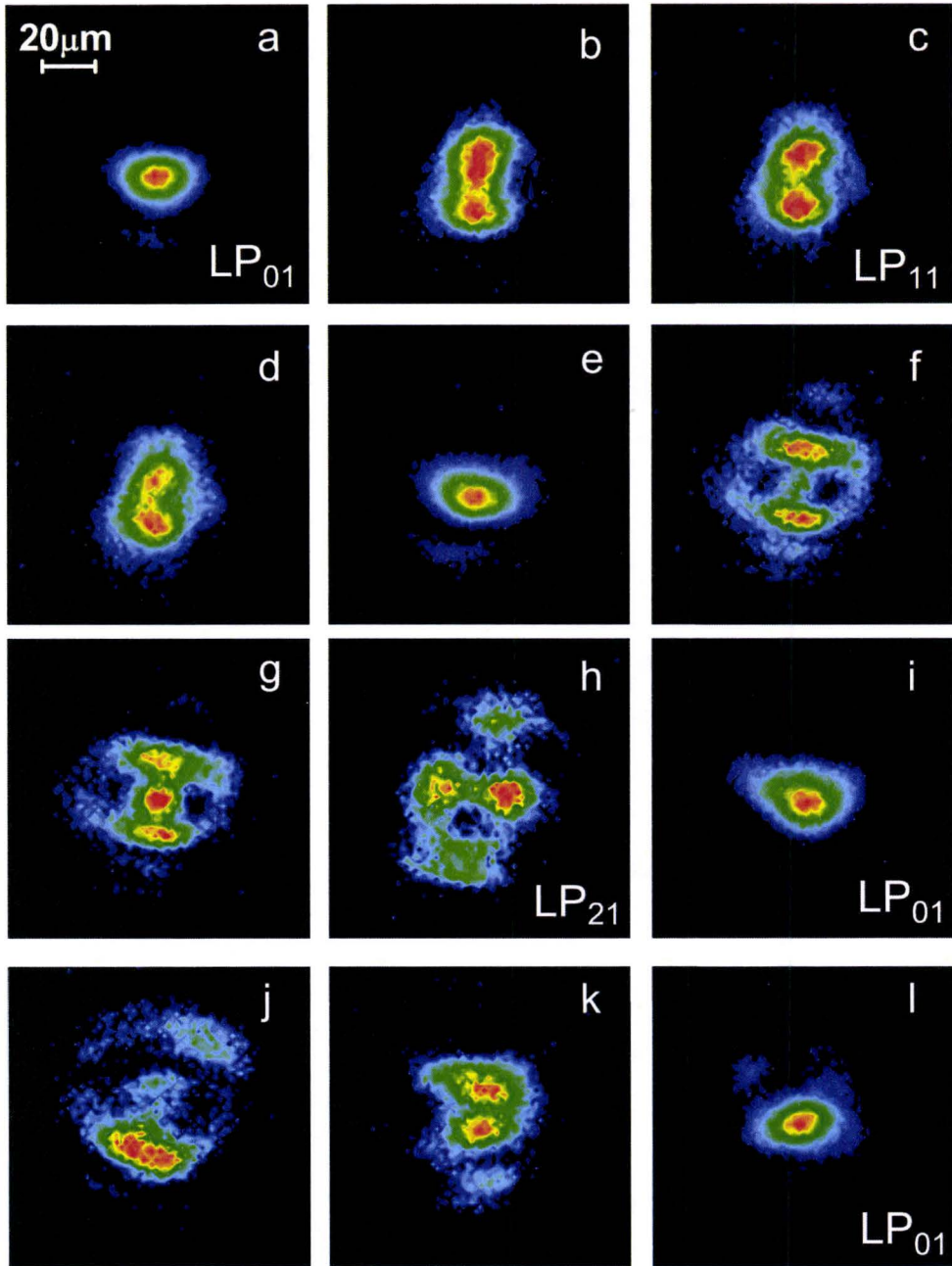


Figure 3.16: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at $3.2 \times 10^{-5} \text{ W/cm}^2$ (average power 0.1 nW). Profiles were acquired at (a) 642 s (b) 671 s (c) 676 s (d) 680 s (e) 720 s (f) 1343 s (g) 1358 s (h) 1381 s (i) 1443 s (j) 1498 s (k) 1531 s and (l) 1552 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

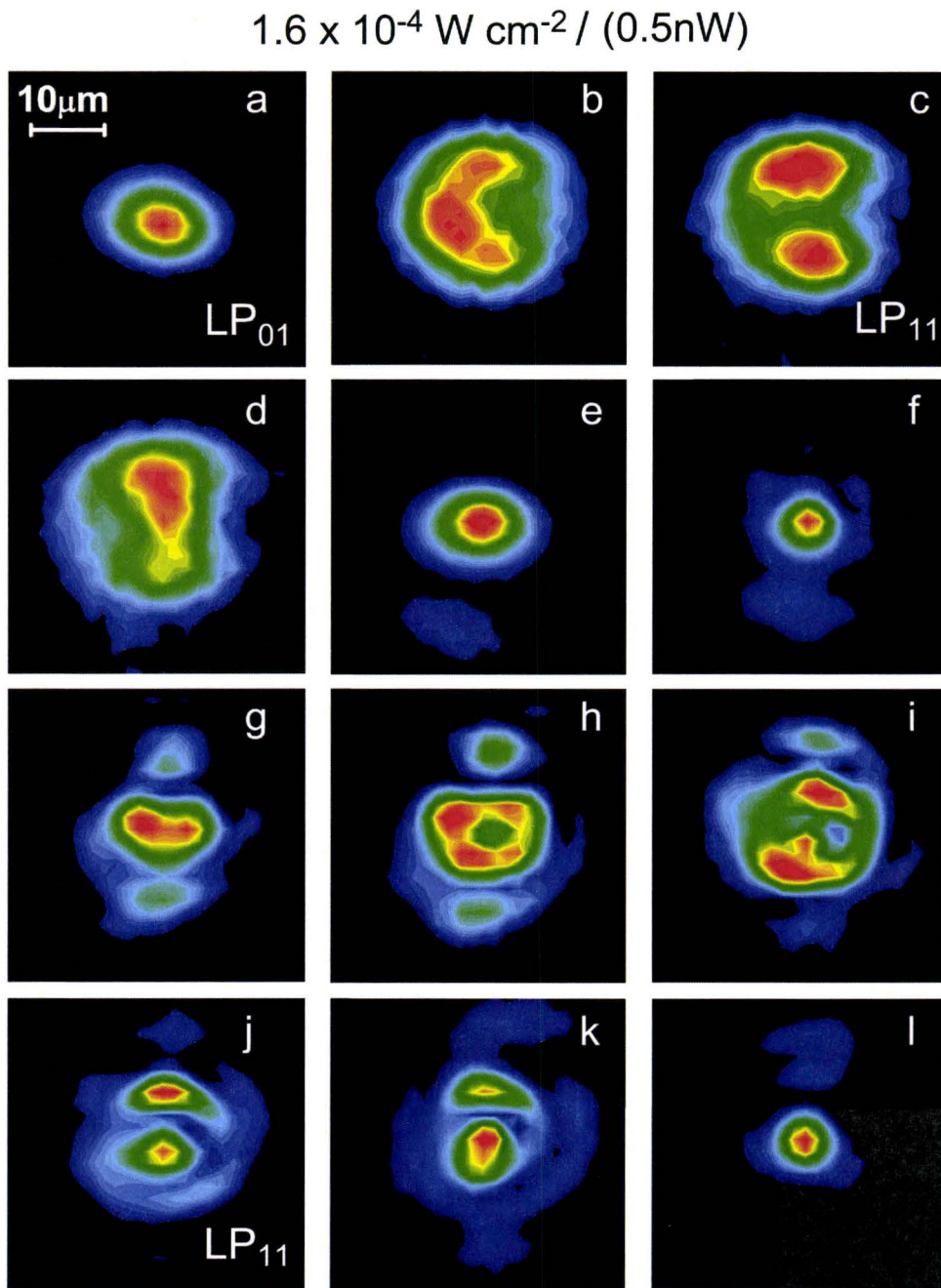


Figure 3.17: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at $1.6 \times 10^{-4} \text{ W/cm}^2$ (average power 0.5 nW). Profiles were acquired at (a) 202 s (b) 214 s (c) 217 s (d) 219 s (e) 242 s (f) 587 s (g) 596 s (h) 600 s (i) 607 s (j) 613 s (k) 618 s and (l) 632 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

$$3.2 \times 10^{-3} \text{ W cm}^{-2} / (10\text{nW})$$

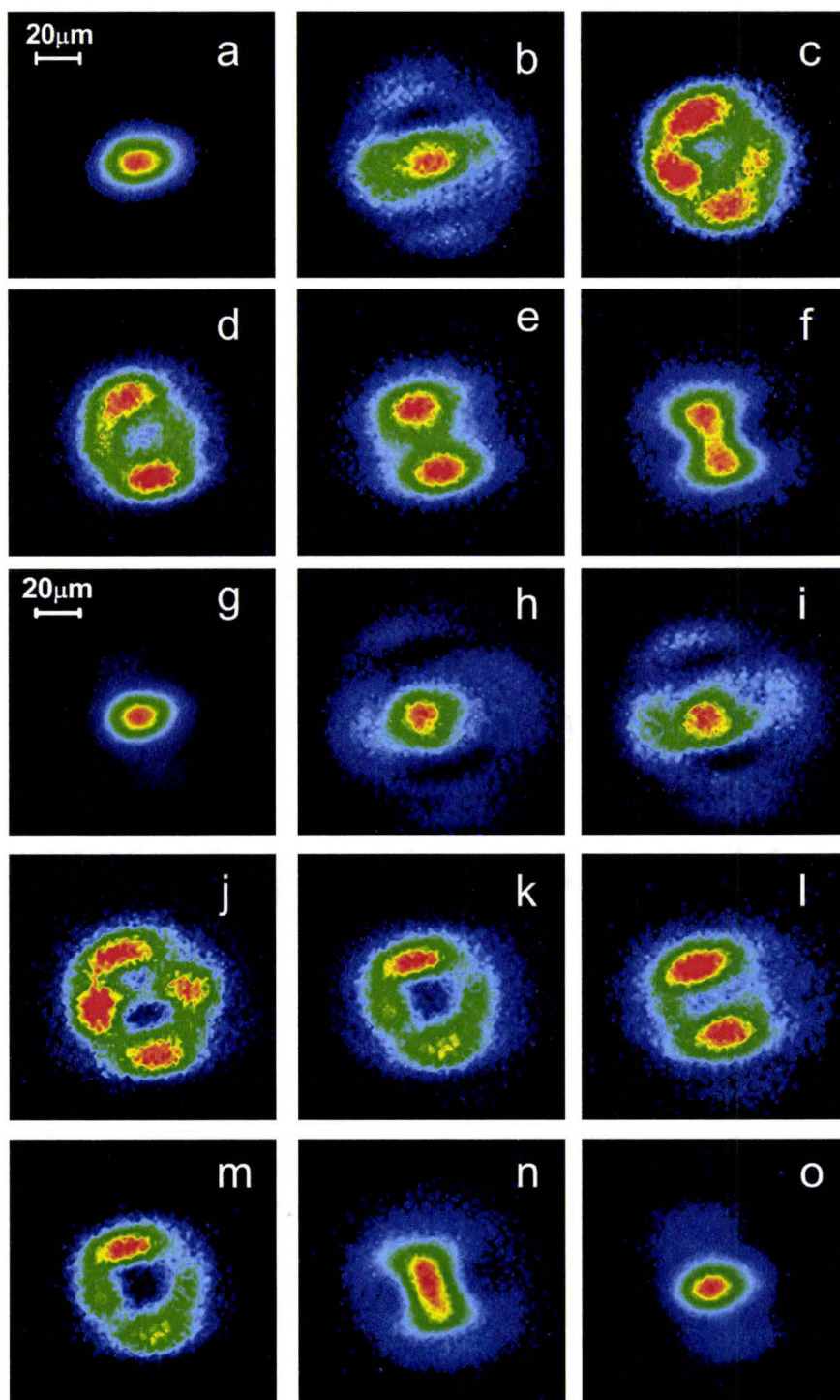


Figure 3.18: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at $3.2 \times 10^{-3} \text{ W/cm}^2$ (average power 10 nW). Profiles were acquired at (a) 105 s (b) 110 s (c) 113 s (d) 114 s (e) 115 s (f) 116 s (g) 119 s (h) 128 s (i) 1297 s (j) 132 s (k) 133 s (l) 134 s (m) 135 s (n) 136 s and (o) 138 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

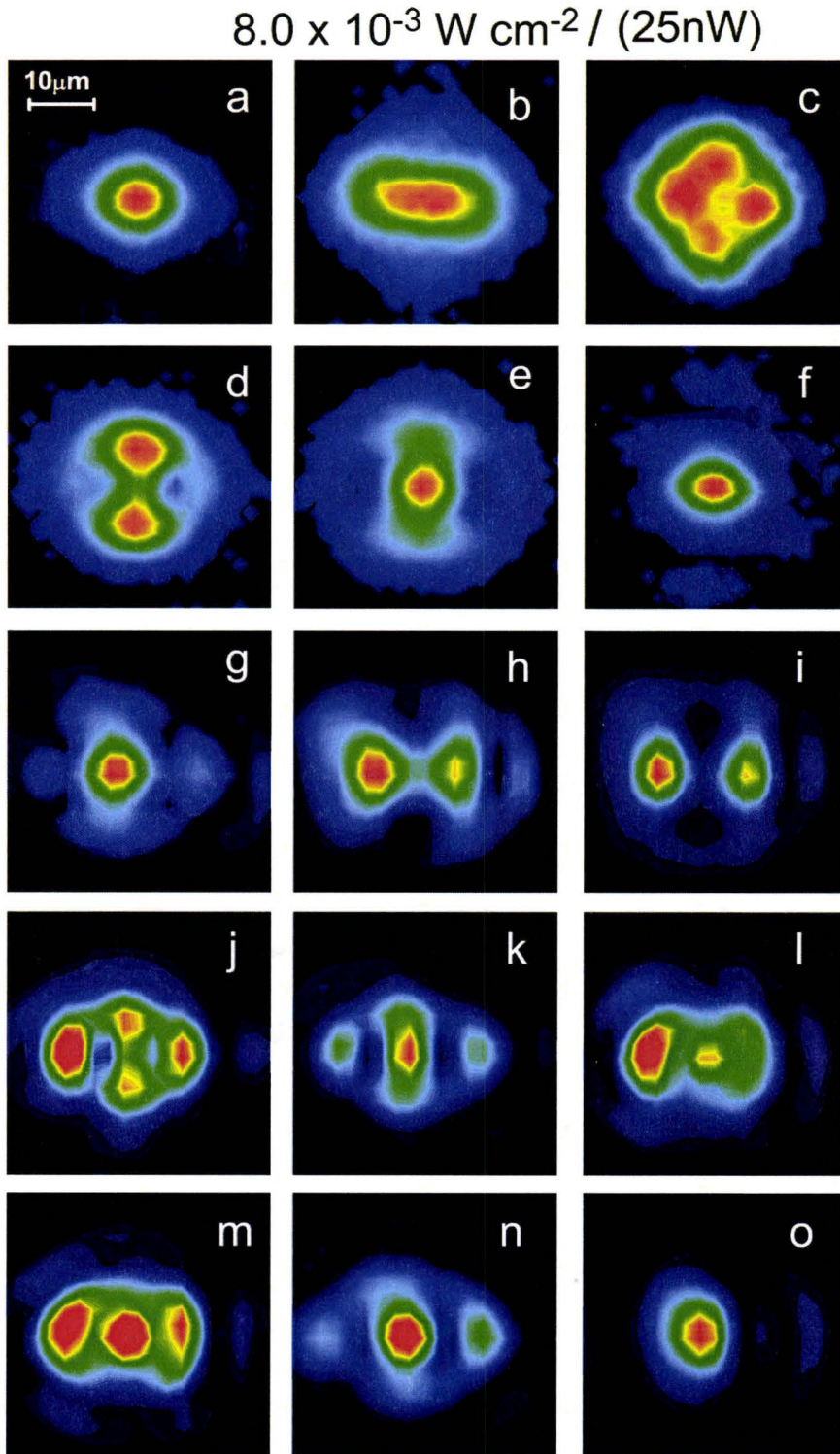


Figure 3.19: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at $8.0 \times 10^{-3} \text{ W/cm}^2$ (average power 25 nW). Profiles were acquired at (a) 19 s (b) 21 s (c) 22 s (d) 23 s (e) 24 s (f) 28 s (g) 74 s (h) 86 s (i) 88 s (j) 93 s (k) 95 s (l) 119 s (m) 121 s (n) 126 s and (o) 134 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

the left of the original beam (at $t = 0$ s). This secondary self-trapping process is probably due to light that had leaked from the (step-index) self-written waveguide and commenced a new self-trapping process in a different (and unpolymerized) region of the medium.

An entirely different phenomenon was observed at 1.6 W/cm^2 the next intensity of the mid-intensity regime; spatial profiles in Figure 3.22 showed the emergence of an increasing number of concentric rings around the beam in the first 14 s. The beam developed a single ring at 4 s (Figure 3.22b) and up to 6 rings were observed in the next 10 s (Figure 3.22h). At 53 s, the beam self-focused and at 160 s (Figure 3.22k) appeared tightly self-trapped with a Gaussian profile and diameter ($1/e^2$) of $20 \mu\text{m}$. The beam remained self-trapped until 219 s, after which it gradually decreased in intensity and increased in width.

These concentric rings have been observed during the propagation of Gaussian beams in other nonlinear optical systems including photorefractive crystals and Kerr media as reviewed in Section 1.3.4 and have been considered both as self-diffracting effects due to spatial self-phase modulation and as dispersive (rather than dissipative) spatial shock waves that bear analogy to shock waves in superfluids [80]. Both cases consider the propagation of a Gaussian beam in a photoresponsive medium (as defined by Equations (3.1) to (3.3)) and relate to the mechanisms that give rise to self-trapping. The formation of diffraction rings constitute a fundamentally different class of nonlinear phenomena, originating from spatial-self-phase modulation which merit separate study and will be further discussed in Chapter 4.

The last intensity investigated in this range, 16 W/cm^2 , showed no evidence of diffraction rings (Figure 3.23, only an initial expansion of the beam at 2 s (Figure 3.23b). Instead, the beam self-trapped within the next 126 s (Figure 3.23h), retained a tightly focused Gaussian profile until 199s (Figure 3.23j), after which it gradually broadened and weakened in intensity.

3.3.3.3 High intensity regime: single ring formation and filamentation

Plots of temporal evolution of beam intensity and width (Figure 3.24) in this regime showed very different behavior compared to the low and mid-intensity regimes. Neither sharp increases in peak intensity nor sudden decreases in whole beam width were observed in the high intensity regime.

3.3.3.4 Single-ring filamentation

At transition intensities between mid and high intensity regimes the beam did not develop multiple diffraction rings or further narrowing of its width, instead we observed the emergence of a single-ring followed by its filamentation. Figure 3.25

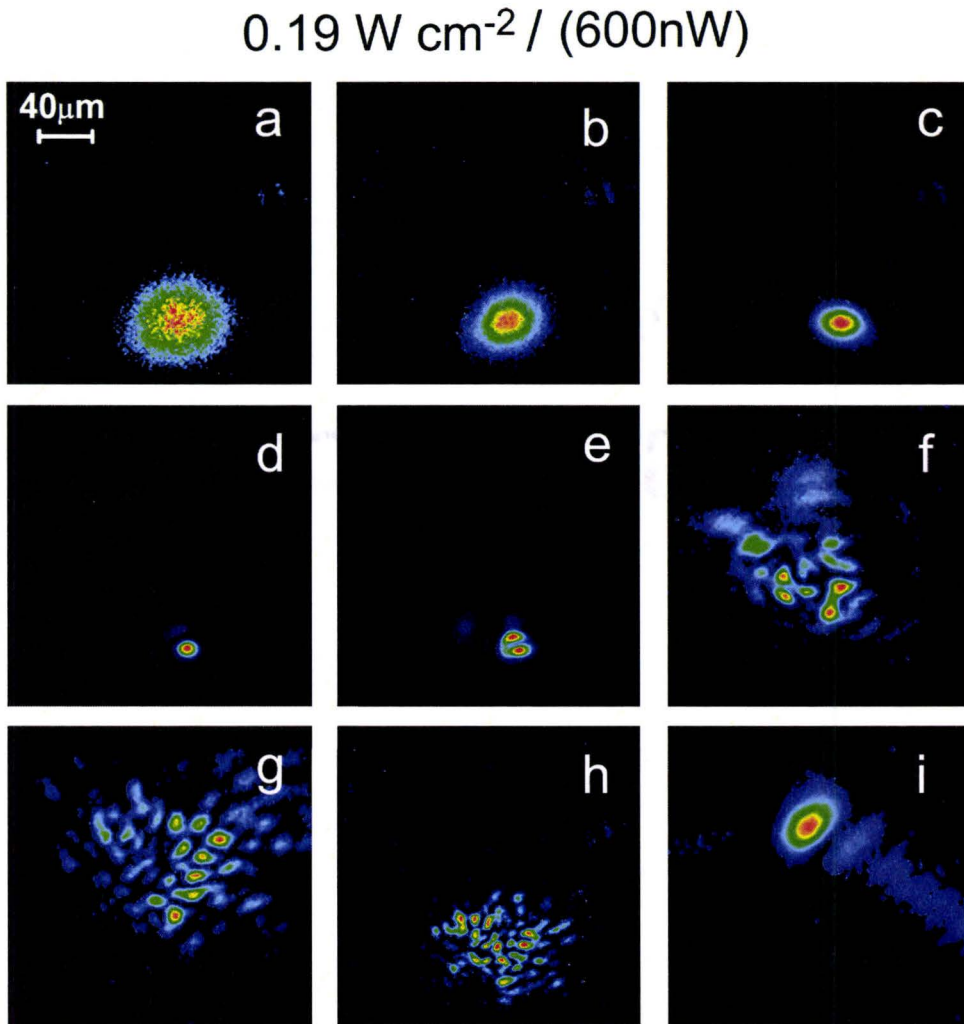


Figure 3.20: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at 0.19 W cm^{-2} (average power 600 nW). Profiles were acquired at (a) 2 s (b) 3 s (c) 6 s (d) 18 s (e) 31 s (f) 57 s (g) 124 s (h) 143 s and (i) 492 s . For clarity, each 2D profile has been normalized to the maximum intensity value.

Mid-intensity regime

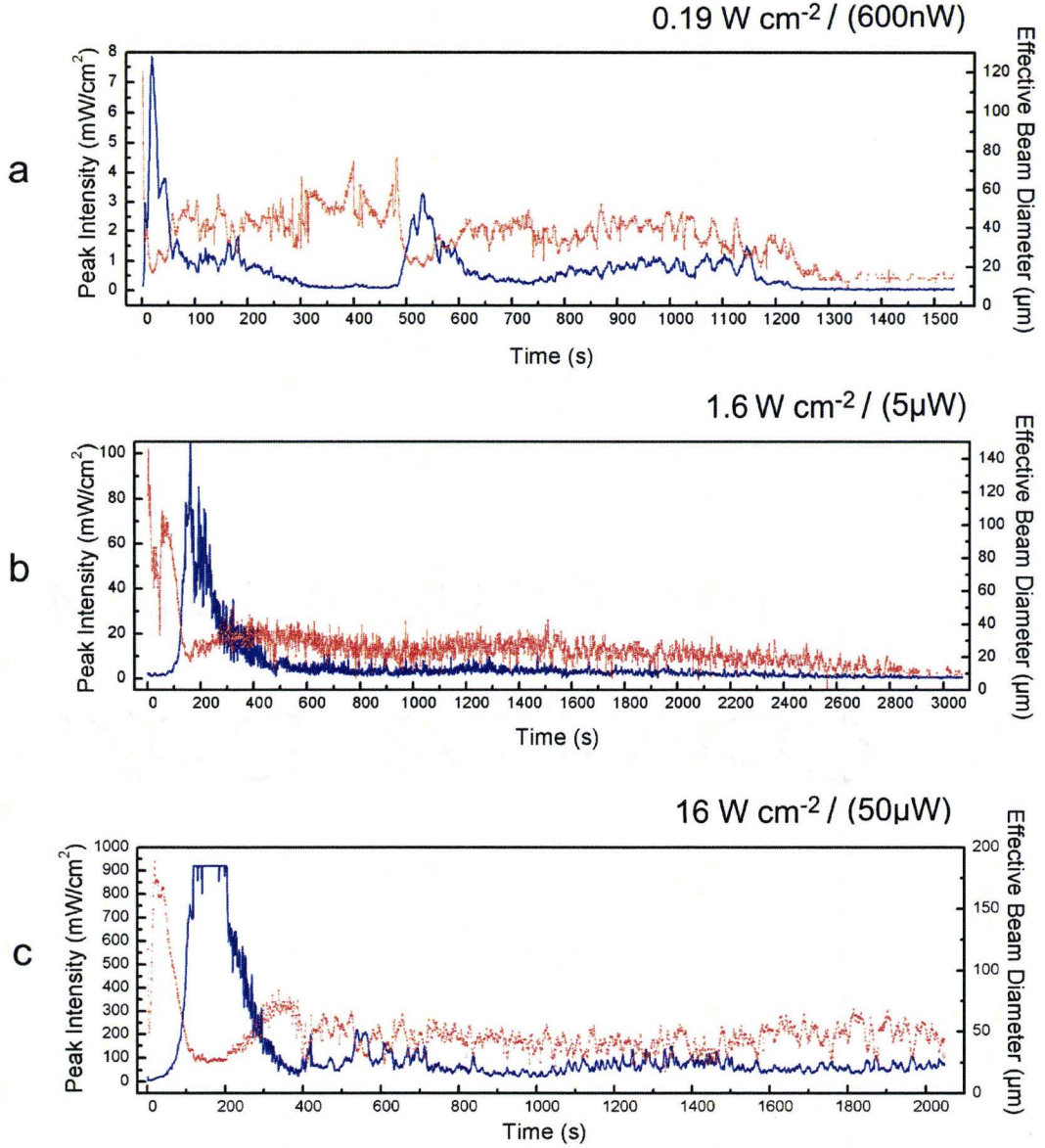


Figure 3.21: Graphs of temporal evolution of peak intensity (solid blue line) and effective beam diameter, $1/e^2$ (dotted red line) collected at the exit face of the organosiloxane corresponding to the mid-intensity regime a) 0.19 W/cm^2 (power 600 nW), b) 1.6 W/cm^2 (power 5 μW) and c) 16 W/cm^2 (power 50 μW).

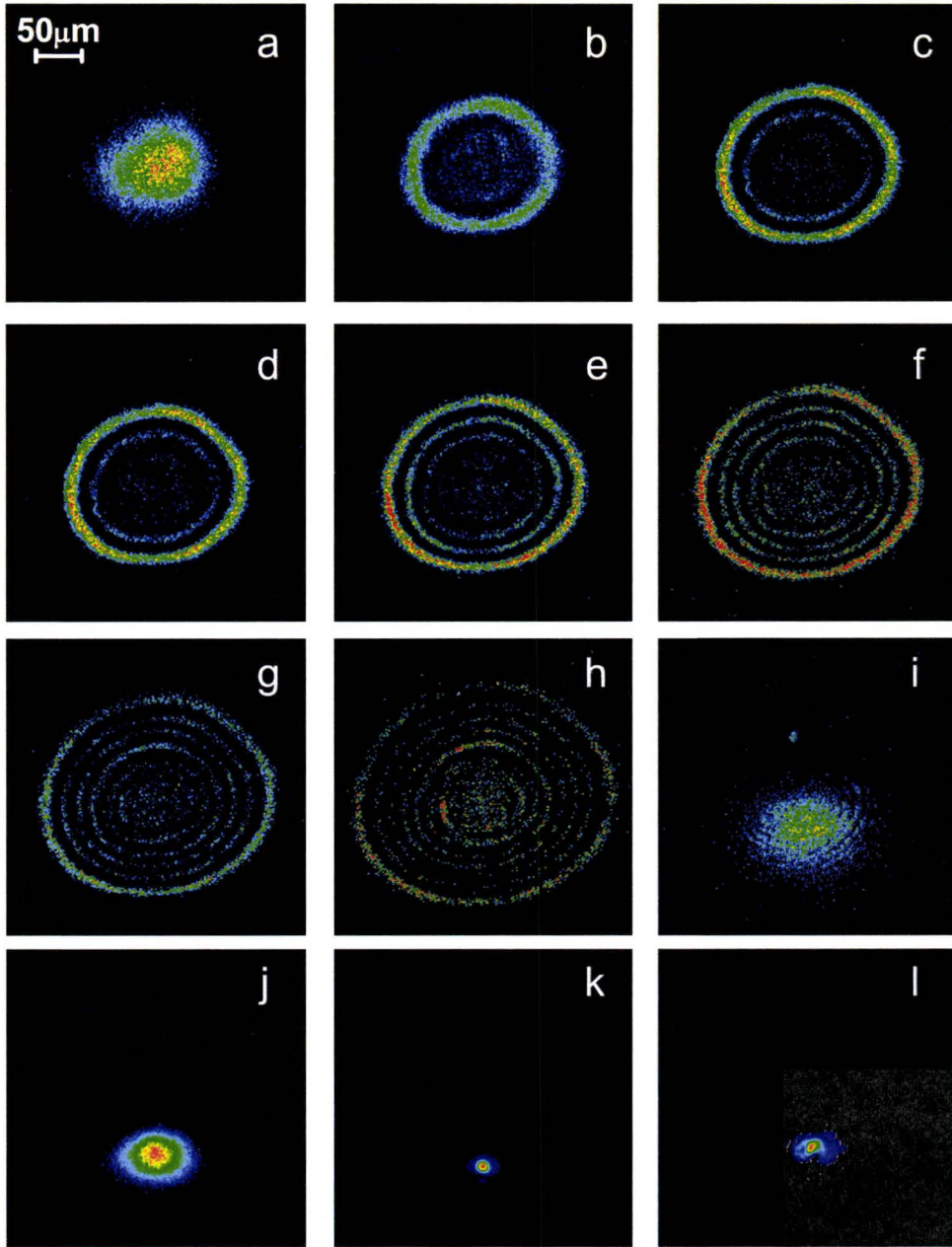


Figure 3.22: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00$ mm) during self-trapping in the organosiloxane at 1.6 W/cm^2 (average power $5\text{ }\mu\text{W}$) in the mid-intensity regime. Profiles were acquired at (a) 1 (b) 4 s (c) 6 s (d) 8 s (e) 9 s (f) 10 s (g) 11 s (h) 14 s (i) 53 s (j) 101 s (k) 160 s (l) 304 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

$16 \text{ W cm}^{-2} / (50 \mu\text{W})$

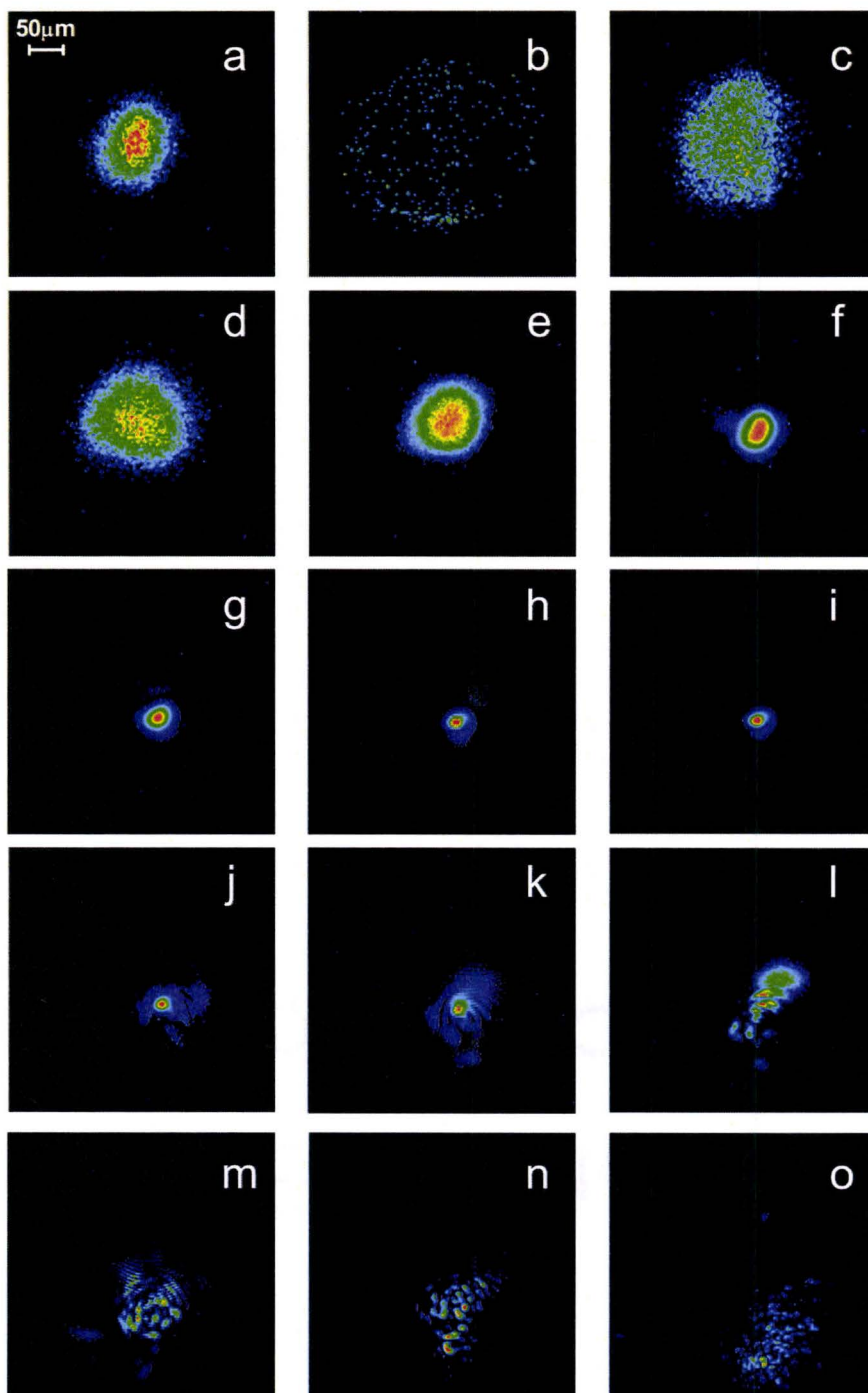


Figure 3.23: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at 16 W/cm^2 (average power $50 \mu\text{W}$). Profiles were acquired at (a) 1 s (b) 2 s (c) 19 s (d) 31 s (e) 55 s (f) 85 s (g) 99 s (h) 126 s (i) 128 s (j) 199 s (k) 255 s (l) 347 s (m) 420 s (n) 767 s and (o) 2045 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

High intensity regime

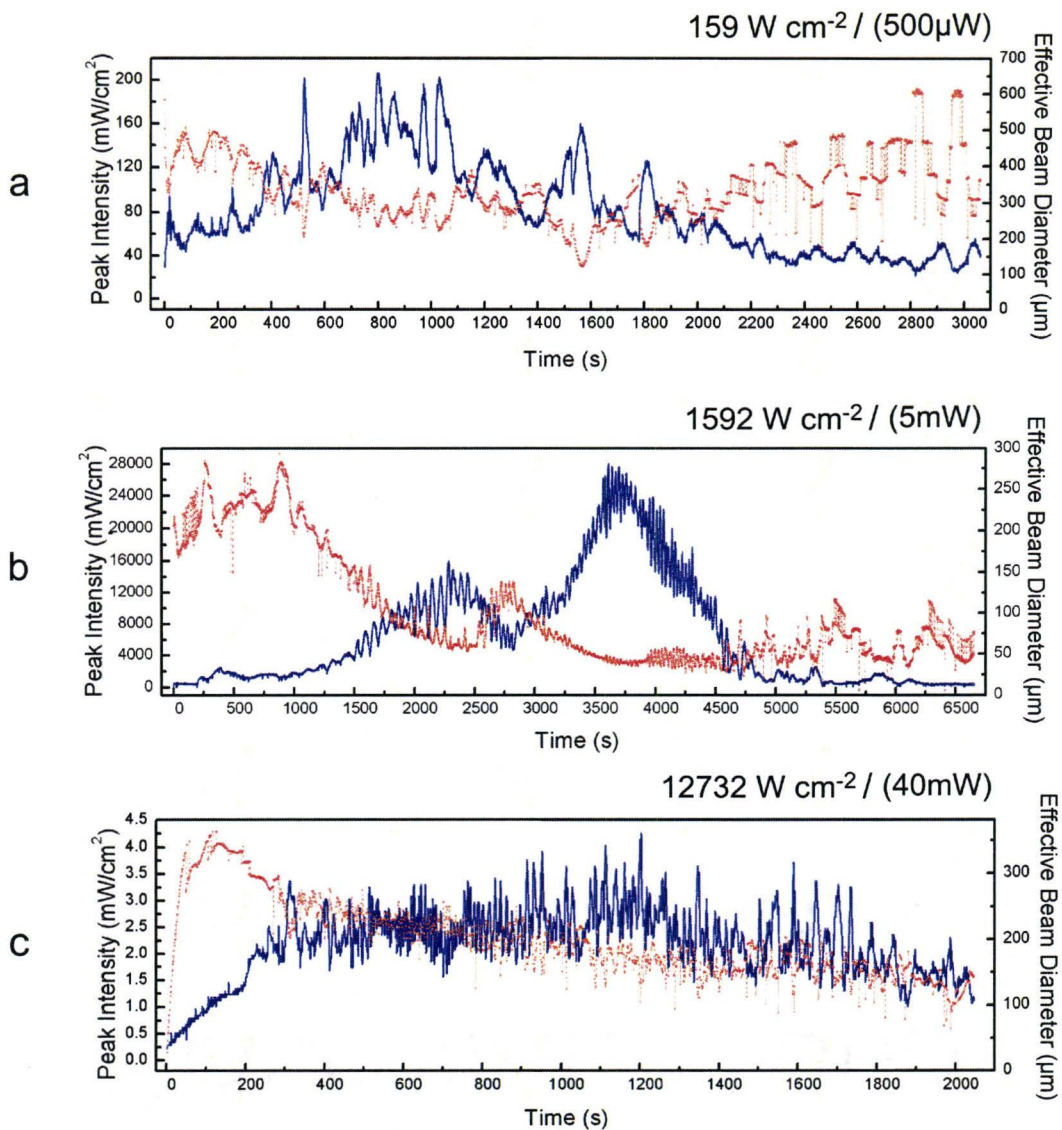


Figure 3.24: Graphs of temporal evolution of peak intensity (solid blue line) and effective beam diameter, $1/e^2$ (dotted red line) collected at the exit face of the organosiloxane corresponding to the high intensity regime: a) 159 W/cm² (average power 500 μW), b) 1592 W/cm² (average power 5 mW) and c) 12732 W/cm² (average power 40 mW).

shows the sequence of 2-D spatial intensity profiles at 159 W/cm^2 , where the beam developed a single-ring with local intensity maxima (Figure 3.25c) that at 325 s (Figure 3.25f) filamented into seven nodes. By 1016 s (Figure 3.25h), the beam had self-divided further into multiple intensity maxima. The corresponding temporal plots of peak intensity and beam width in Figure 3.24a, show a broad peak in intensity at around 800s, that occurred later compared to the fast rising and narrow peak intensity in the mid-intensity regime. The broad peak corresponds to the increase in intensity during filament formation but no self-trapping of the whole beam was observed.

The phenomenon of single-ring formation accompanied with filamentation has been observed in Kerr media for input Super-Gaussian (SG) beams as reviewed in Section 1.4.3. In the case of the organosiloxane, the saturation of the refractive index induces a flattened Gaussian index profile (similar to SG beams), which gives rise to the single-ring observed in our experiments. In Section 3.3.1 we will discuss in more detail the role of the saturation of the refractive index profile and in Chapter 5 we will further investigate the origin of single-ring formation and its filamentation dynamics over a range of intensities.

3.3.3.5 Whole beam filamentation

At the greatest intensities, including 1592 W/cm^2 and 12732 W/cm^2 we observed splitting and filamentation of the whole beam

At 1592 W/cm^2 , the beam divided into two segments which later self-trapped at different times (Figure 3.26). In the corresponding temporal plot (Figure 3.24b), the self-trapping of each segment was observed as two distinct regions of increasing intensity and decreasing width. No individual self-trapped beams were observed at 4607 s (Figure 3.26h), by which point the beam had developed multiple intensity maxima.

At the greatest intensity of 12732 W/cm^2 , the beam was observed to filament due to rapid variations (Figure 3.27), possibly originating from thermal convection in the organosiloxane. At 218 s, the beam split in two parts (Figure 3.27e) and subsequently into four parts at 413 s (Figure 3.27h) until it developed multiple filaments at 1476 s (Figure 3.27k). Temporal plots of intensity and beam diameter in Figure 3.24c showed only an initial increase in intensity in the first 200 s, followed by a steady value in intensity and beam width (only small fluctuations) over the next 1400 s. The initial increase in intensity is probably due to an increase in refractive index over the whole area of the beam.

Filamentation of the beam occurs through a mechanism similar to modulation instability and spontaneous division of uniform beams in polymerizable media (see Section 1.4). In the high-intensity regime, the induced refractive index gradient is not sufficient to collectively self-trap the entire Gaussian beam (see Sections 3.3.1

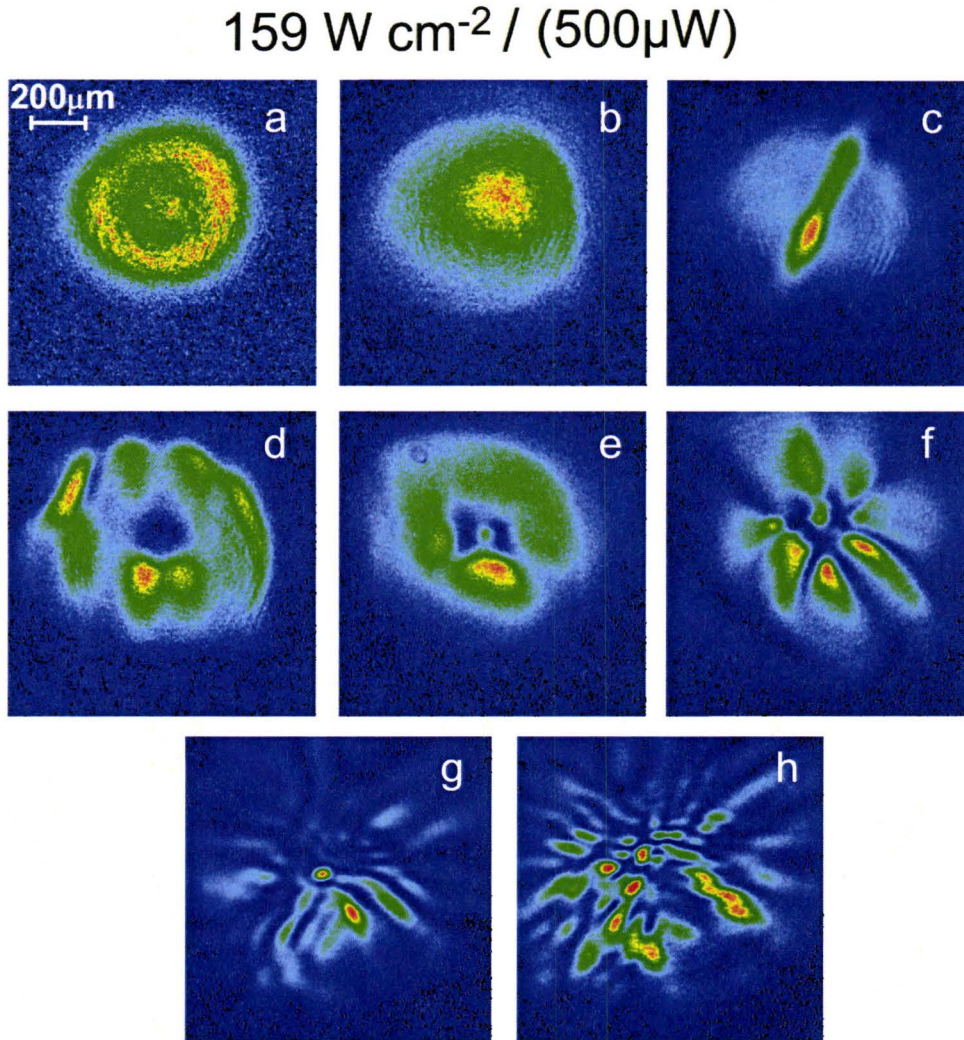


Figure 3.25: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at 159 W/cm^2 (average power $500 \mu\text{W}$) in the high intensity regime. Profiles were acquired at (a) 1 s (b) 3 s (c) 19 s (d) 43 s (e) 108 s (f) 325 s (g) 798 s and (h) 1016 s . For clarity, each 2D profile has been normalized to the maximum intensity value.

$1592 \text{ W cm}^{-2} / (5\text{mW})$

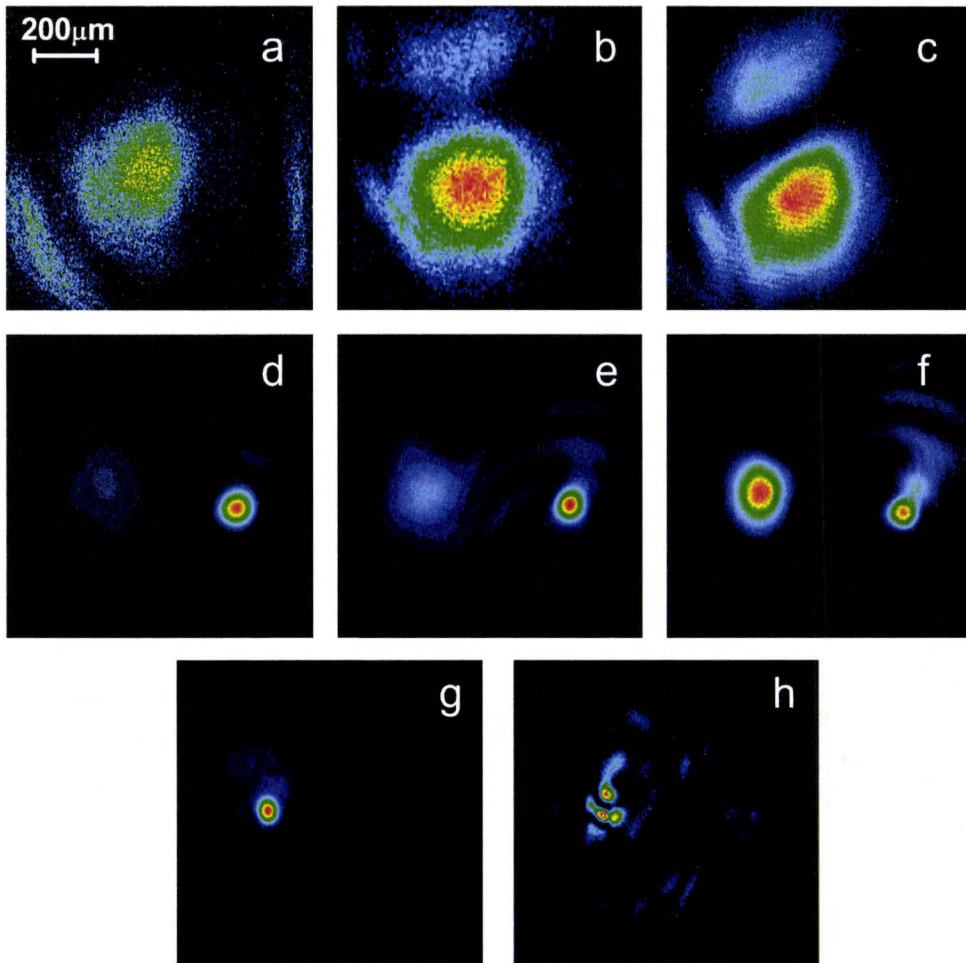


Figure 3.26: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00 \text{ mm}$) during self-trapping in the organosiloxane at 1592 W/cm^2 (average power 5 mW) in the high intensity regime. Profiles were acquired at (a) 1 s (b) 233 s (c) 444 s (d) 2285 s (e) 2558 s (f) 2814 s (g) 3614 s and (h) 4607 s . For clarity, each 2D profile has been normalized to the maximum intensity value.

and 3.3.2). However, noise imposed on the optical beam - such as minor variations in refractive index of the medium or beam intensity - become amplified under nonlinear conditions and ultimately disintegrate into individual self-trapped filaments. At 159 W/cm^2 , intensity variations within the single-ring at 43 s, seeded fragmentation (Figure 3.25d). At the greater intensities of 1592 W/cm^2 and 12732 W/cm^2 , the sequence of fragmentation was random and probably relies on random fluctuations in intensity and/or refractive index. These findings are consistent with previous observations of the spontaneous disintegration of a beam into multiple filaments and the self-inscription of multiple self-written waveguides [58; 59].

3.4 Conclusions

In this Chapter, we provided a comprehensive overview of the nonlinear propagation of a continuous-wave Gaussian laser beam in a medium undergoing photopolymerization over 10 order of magnitude in intensity. Figure 3.28 presents a summary of the different observed phenomena at different intensities. These results confirmed and provided new insight into key predictions of theoretical models developed in the past decade, such as the excitation of high-order optical modes of the self-trapped beam and concomitant self-inducement of a multimode waveguide. Most significantly, these experiments provided an extremely rare opportunity to directly observe the excitation of individual higher order modes during the evolution of a cylindrical waveguide from single-mode to multimode guidance. Trends in the oscillatory behavior of the modes during this process were consistent with simulations performed and those predicted by theory. Quantitative analyses of self-trapping at a broad range of intensities revealed the significance of the gradient of photo-induced refractive index changes, showing that self-trapping was most efficient in the low-intensity regime where the steepest gradients are induced. While beam filamentation dominated at extremely high intensities, new forms of nonlinear propagation in photopolymerizable media including spatial diffraction rings and single ring formation were observed in the mid-intensity regime and close to the high intensity regime, respectively. Chapter 4 will present the study of the dynamics of diffraction rings in the organosiloxane and Chapter 5 will provide a detailed study of single ring formation.

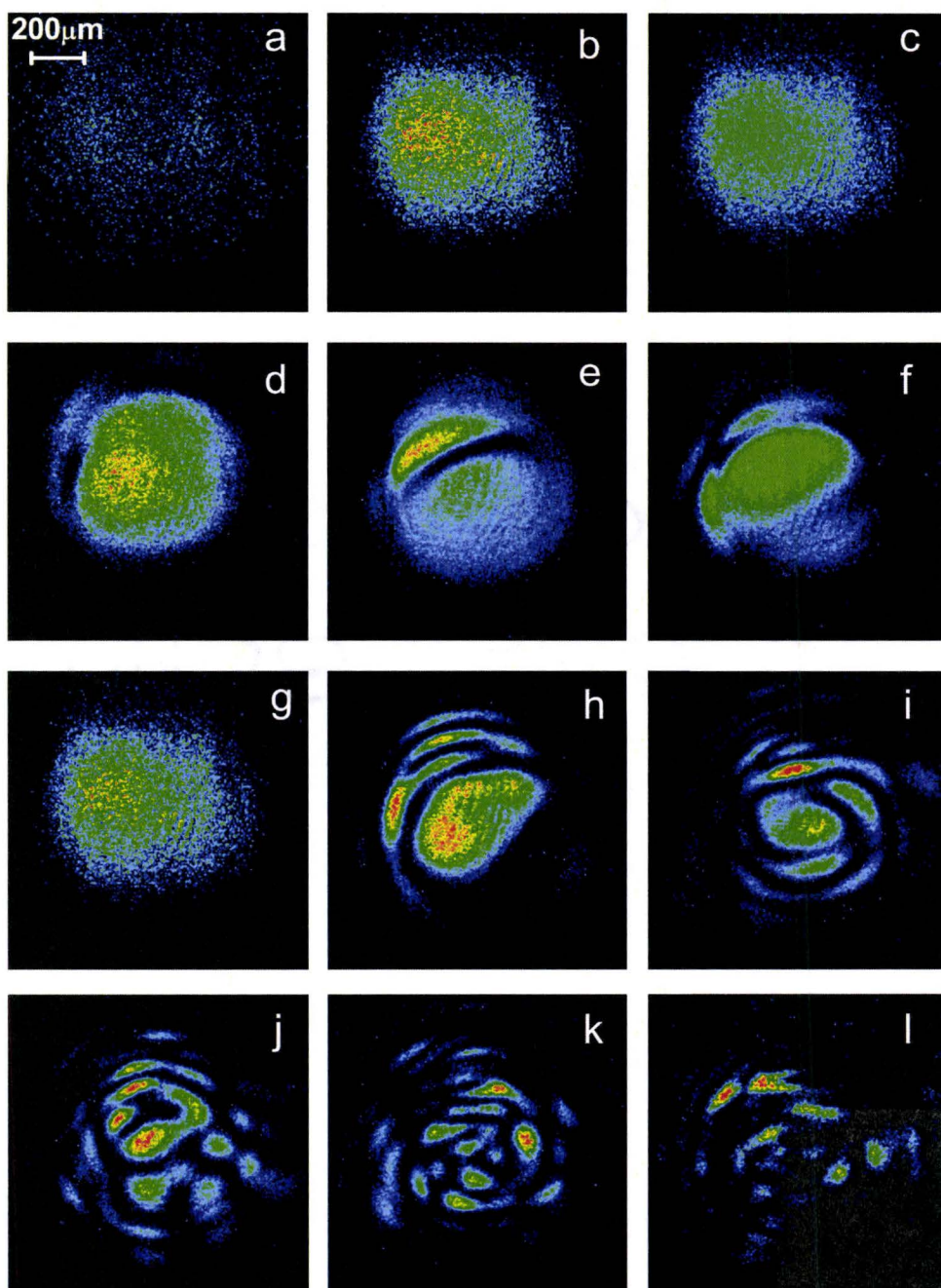


Figure 3.27: Temporal variations of 2-D spatial intensity profiles of the beam at the exit face ($z = 6.00$ mm) during self-trapping in the organosiloxane at 12732 W/cm^2 (average power 40 mW). Profiles were acquired at (a) 20 s (b) 65 s (c) 147 s (d) 160 s (e) 218 s (f) 254 s (g) 255 s (h) 413 s (i) 767 s (j) 1022 s (k) 1476 s (l) 2046 s. For clarity, each 2D profile has been normalized to the maximum intensity value.

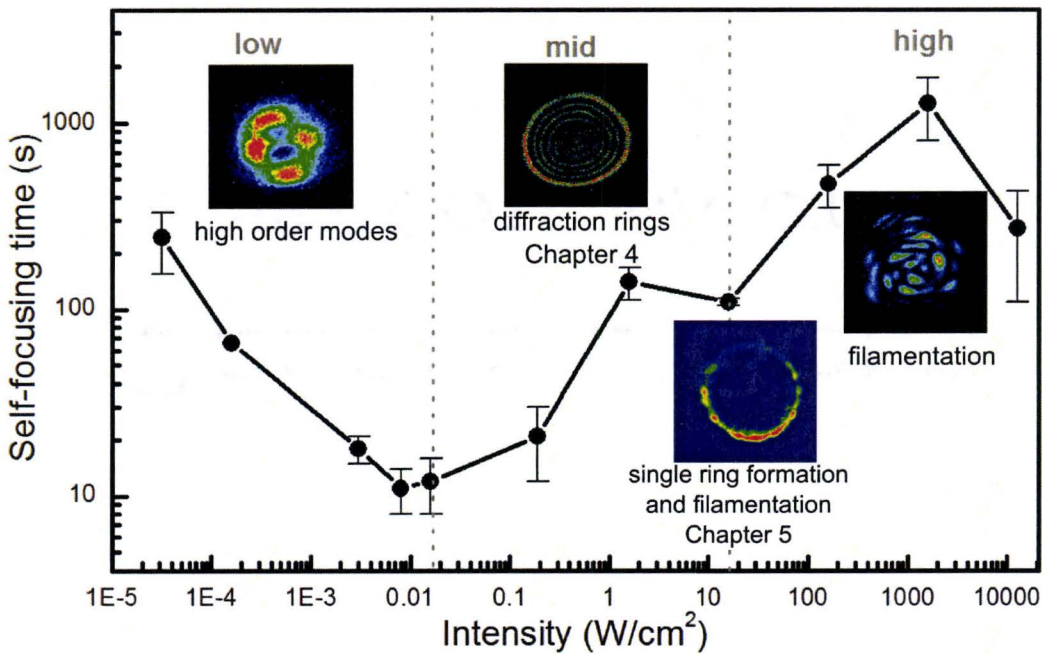


Figure 3.28: Representation of the four different forms of nonlinear light propagation in the photopolymerizable organosiloxane in the three intensity regimes. The low intensity regime is characterised by the modal evolution of the self-trapped beam while self-diffraction rings and beam filamentation occur in the mid and high-intensity regimes, respectively. Between these two intensity regimes single ring formation with further filamentation occurs.

Chapter 4

Diffraction rings by spatial self-phase modulation

4.1 Introduction

Nonlinear propagation of a continuous wave visible laser in the organosiloxane was studied at a range of intensities spanning 10 orders of magnitude. In this Chapter, the mid-intensity regime with intensities ranging from 1.6 to 16 W/cm² is further explored. This intensity regime revealed the emergence of diffraction rings due to spatial self-phase modulation in the organosiloxane. To our knowledge this is the first example of spatial self-phase modulation (Section 1.3) originating from laser initiated free-radical polymerisation and corresponding changes in the refractive index of the medium. However, this study focuses on the *differences* in the photoresponse of the organosiloxane relative to other nonlinear optical materials and the opportunities that they provide to probe previously inaccessible properties and dynamics of the self-induced diffraction rings.

Specifically, the noninstantaneous response of the organosiloxane enabled diffraction rings to propagate through long distances ($\gg$ Rayleigh length (z_R)) in the medium without disruption from optical self-focusing. It was moreover possible to monitor the temporal evolution of the rings for different organosiloxane pathlengths and thereby, gain direct insight into the dynamics of self-phase modulation. Variation in the input beam gave insight into new types of propagations and resulting intensity profiles for photopolymers. Furthermore, refractive index changes due to polymerisation were permanent and provided a direct view of the conical trajectory of the diffraction rings through the medium.

As we have shown in previous studies, photoinduced changes in the refractive index of organosiloxane originate from a chemical reaction, namely photoinitiated free-radical polymerization of methacrylate substituents [27; 106; 21; 24; 25]. This contrasts with nonlinear media previously employed to study self-phase modulation, in which index changes arise from high-order dielectric susceptibilities as in the cases of atomic vapours [5; 87; 86; 77], Kerr media [75; 83] and photorefractive crystals [80], the Freedericksz transition as in the case of nematic liquid crystals [6; 78; 82] or are thermally induced [73; 74; 4; 76] (Section 1.3). Fundamental differences therefore exist between these photophysical mechanisms and the photochemical response of

organosiloxane (Section 1.3.4). For example, in contrast to the virtually instantaneous photoresponse of Kerr media and atomic vapours, the rate of refractive index changes caused by photopolymerization is slow, spanning milliseconds to seconds [27; 106; 21].

Under these conditions, we find that self-phase modulation is not disrupted by simultaneous self-focusing of the beam, which enables diffraction rings to propagate over long distances ($\gg$ Rayleigh range (z_R)) within the medium. This contrasts with the majority of studies of self-phase modulation, which had to be carried out in thin samples (with pathlengths $< z_R$) in order to prevent simultaneous self-focusing of the beam. Another exception is the recent observation of diffraction rings that propagate over long distances due to a self-defocusing nonlinearity in a photorefractive crystal [80]. The slow photoresponse of the organosiloxane also enables the temporal evolution of diffraction rings to be monitored *in situ* and in this way, provides insight into the dynamics of the self-phase modulation process. Furthermore, because photopolymerisation is an irreversible chemical reaction, index changes induced by the beam are permanently imprinted in the organosiloxane [21]. The resulting microstructures induced by the diffraction rings therefore do not decay in the absence of the optical field and provide an unambiguous view of their trajectory through the medium.

Moreover, through a study of the effect of beam curvature (R) we found the propagation of two types of rings, rings with a dark central spot could be induced by a convergent beam ($R < 0$) while rings with a bright central spot were induced by a divergent beam ($R > 0$). Detailed analyses of the statistics of diffraction rings induced at 1.6 W/cm^2 at different beam curvatures and propagation pathlengths were also carried out. Different types of diffraction rings elicited at $R = \infty$ including rings with high-order modes and fingerprint rings were also identified. Through a pathlength dependence study we found the different diffraction ring dynamics through the organosiloxane.

4.2 Self-phase modulation due to laser initiated free-radical polymerization

In Chapter 3 we demonstrated that the kinetics of photopolymerisation and consequently, the spatial and temporal evolution of refractive index changes strongly influence the type of nonlinear phenomenon that can be elicited in the organosiloxane. When the rate of polymerisation was controlled by varying the incident optical intensity across 10 orders of magnitude ($3.2 \times 10^{-5} \text{ W/cm}^2$ to 12732 W/cm^2), a c.w. 532 nm beam propagating in a photopolymerisable organosiloxane exhibited three distinct forms of behaviour. Optical self-trapping including the sequential excitation of high order modes in the self-trapped beam dominated the low intensity-regime ($3.2 \times 10^{-5} \text{ W/cm}^2$ to 0.16 W/cm^2) whereas single ring formation and filamentation of the beam due to modulation instability occurred in the high intensity-regime

(159 W/cm² to 12732 W/cm²). The diffraction rings described in this Chapter were elicited selectively in the mid-intensity regime (1.6 W/cm² to 16 W/cm²) [21].

Preparation of the organosiloxane and the optical assembly employed in the current study have been described in detail in Sections 2.2 and 2.3 respectively. Briefly, the medium consisted of a sol of organosiloxane oligomers with photopolymerizable methacrylate substituents, which was sensitized to visible wavelengths ($\lambda_{max} = 393, 460$ nm) through addition of 0.05 wt% of a titanocene free-radical photoinitiator. The sol was contained in a cylindrical cell ($d = 12$ mm, pathlength = 6 mm) with optically flat, transparent windows. Samples were partially polymerized and transformed into gels through uniform illumination with white light from an incandescent quartz-tungsten-halogen lamp. Diffraction rings were induced by a linearly polarized, continuous wave 532 nm beam that was focused to a diameter ($1/e^2$) of 20 μ m onto the entrance face of the organosiloxane gel. The spatial intensity profile of the beam at the exit face was imaged onto a charge-coupled device camera and monitored over time. Using an approximate value of 1.47 for the refractive index of the medium [24], z_R and optical path length (OPL) of the beam were calculated to 0.87 mm and 8.82 mm, respectively.

Figure 4.1 contains the spatial intensity profile of diffraction rings observed at the exit face of the organosiloxane medium. The rings were induced at an input laser intensity of 1.6 W/cm²; under these conditions a maximum of 13 rings were observed after 37 s. Diffraction rings in the organosiloxane originate from spatial self-phase modulation, the theory of which was initially described by Dabby and coworkers [74] and developed by Durbin [6], Le Berre [81], Santamato [82], Deng [85] and coworkers (For details see Section 1.3).

Self-phase modulation in the organosiloxane originates as follows: initially, the optical field intensity and therefore, the photoinduced refractive index change are greatest at the entrance face. Due to the excellent spatial resolution in the photoresponse of the organosiloxane (~ 150 nm) [25], the spatial profile of this index change closely corresponds to that of the 20 μ m-wide Gaussian beam (as previously confirmed through beam propagation simulations)[21]. The modified index profile imposes a transverse phase shift over the entire beam expressed as

$$\Delta\psi(r) = \frac{2\pi}{\lambda} \int_{z_0}^{z_0+d} \Delta n(r, z) dz \quad (4.1)$$

where $\Delta n(r, z)$ is the induced change in refractive index at a specific point in space, λ is the wavelength of the optical field in free-space, z_0 represents the entrance face along the propagation axis (z) and d is the propagation distance along which the beam acquires a transverse phase shift [6]. $\Delta n(r, z)$ in the organosiloxane is given by the empirical formula derived by Kewitsch and Yariv [20]

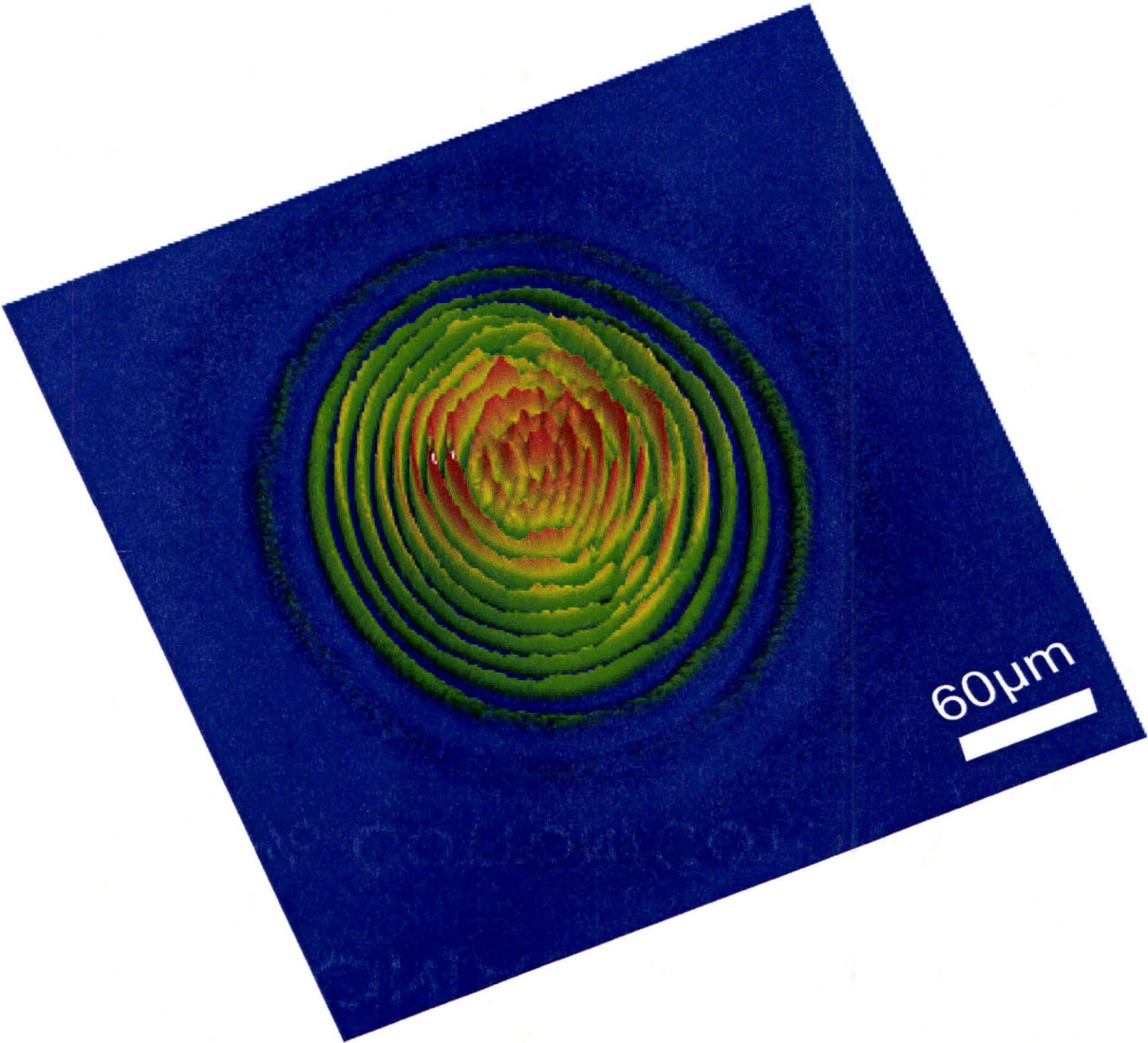


Figure 4.1: Intensity profile of diffraction rings at the exit face of the organosiloxane induced with an input laser intensity of 1.6 W/cm^2 .

$$\Delta n(x, y, z, t) = \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(t)|^2 dt \right] \right\} \quad (4.2)$$

where Δn_s is the maximum refractive index change, U_0 is the critical exposure required to initiate polymerization, τ is the monomer radical lifetime and $E(t)$ is the amplitude of the electric field. In Equation (4.2), an input Gaussian beam can induce a refractive index change with the same profile due to the intensity dependence of $\Delta n(r, z)$. The resulting transversal phase shift profile in the organosiloxane is given by substitution of Equation (4.2) in Equation (4.1)

$$\Delta\psi(r) = \frac{2\pi}{\lambda} \int_{z_0}^{z_0+d} \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(r, z, t)|^2 dt \right] \right\} dz \quad (4.3)$$

here the integrand with respect to z corresponds to the refractive index change in the photopolymer. According to Equation (4.3), for an input Gaussian beam, the profile of the phase shift $\Delta\psi(r)$ must correspond to the Gaussian profile of the refractive index change Δn for intensities below saturation of the refractive index. Radiation from any two points (r_1, r_2) along $\Delta\psi(r)$ that have the same slope, and thereby the same wavevector, $k_\perp = (d\Delta\psi(r)/dr)$, suffer interference when $\Delta\psi(r_1) - \Delta\psi(r_2) = m\pi$. Constructive or destructive interference occurs when m is, respectively, an even or odd integer with multiple rings forming when the maximum phase shift $\Delta\psi_0 > 2\pi$. As the refractive index increases in the organosiloxane, the maximum value of $\Delta\psi(r)$ increases and more points having the same wavevector are available to interfere, creating an increasing number of rings. Interference coupled with the cylindrically symmetric profile of $\Delta\psi(r)$ produces an array of interference cones, which when projected onto a plane at $z \gg z_R$ are observed as bright and dark concentric diffraction rings (Figure 4.1). The outermost ring originates from radiation with the greatest $k_\perp$ about the inflection point, $(d\Delta\psi(r)/dr)_{max}$, and is therefore initially the most intense [6] (Figures 4.2 (a)-(e)).

4.3 Propagation of diffraction rings over long distances ($\gg z_R$)

The diffraction rings presented in Figure 4.1 were observed at the exit face of the organosiloxane after an optical pathlength (OPL) of 8.82 mm. This contrasts sharply with most previous studies where self-phase modulation could be induced only in thin samples and diffraction rings, observed in the far field after propagation through air [74; 4; 76; 6]. In these cases, samples of thickness $< z_R$ were employed in order to suppress simultaneous self-focusing of the beam that would otherwise disrupt self-phase modulation. This problem is entirely avoided in the organosiloxane due to its noninstantaneous response [21], which delays the onset of self-focusing and enables the

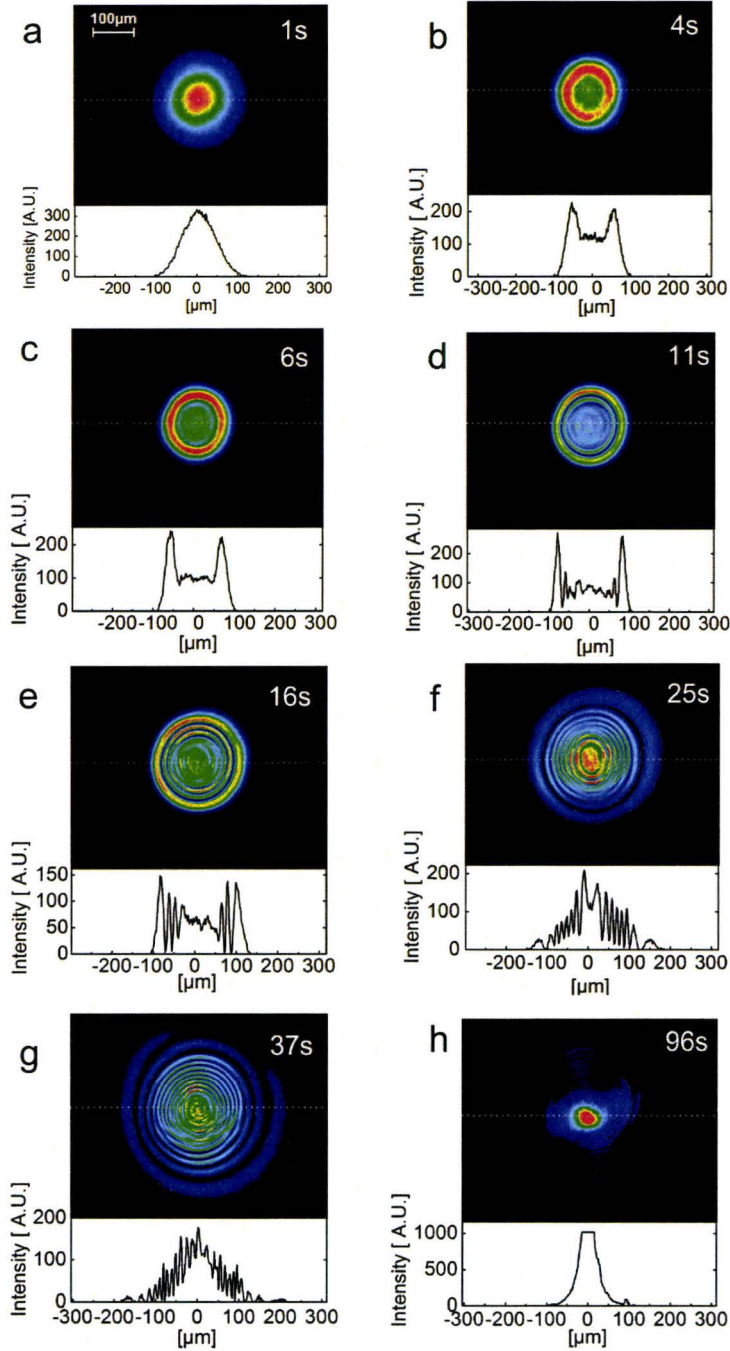


Figure 4.2: Temporal evolution of diffraction ring formation at an intensity of 1.6 W/cm^2 in the organosiloxane with $OPL = 8.82 \text{ mm}$. 2-D and 1-D spatial intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value.

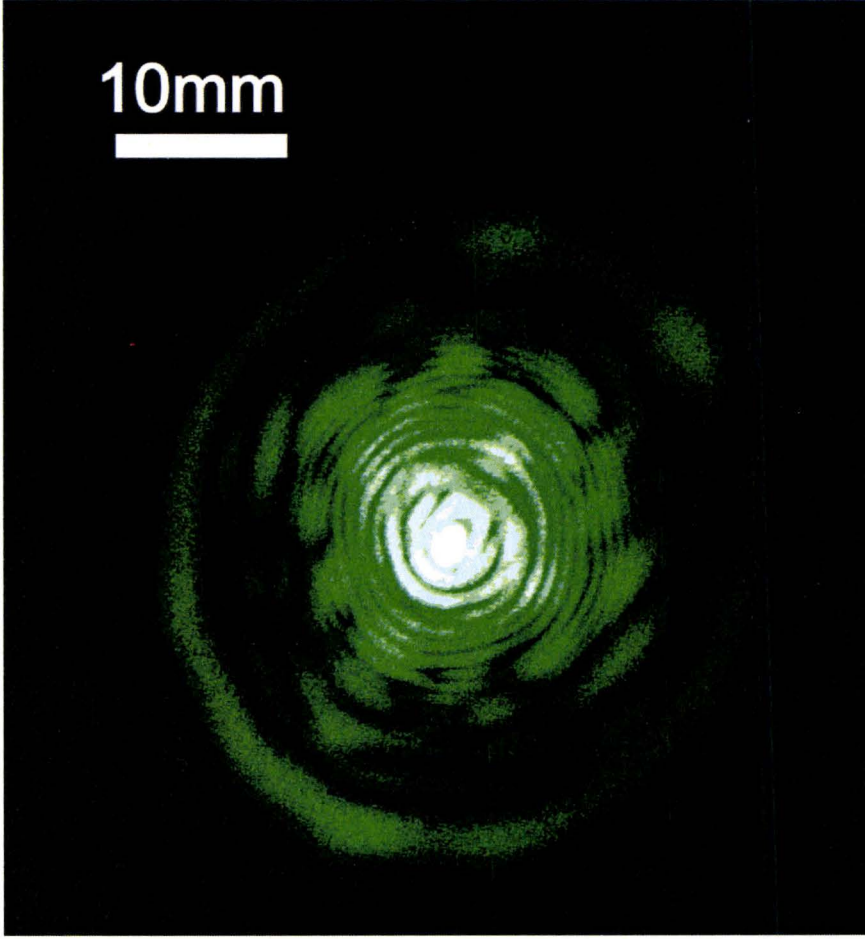


Figure 4.3: Photograph of diffraction rings elicited in a thin sample (0.56 mm thick) with $\text{OPL} = 0.82$ mm at 1.6 W/cm^2 projected onto a screen in the far field.

period of self-phase modulation (≈ 40 s) to be clearly distinguished from subsequent self-focusing of the beam (Figures 4.2 (a)-(g)). The appearance of diffraction rings at $\text{OPL} \gg z_R$ *within* the organosiloxane medium itself can be explained as follows: at early times, the refractive index change and consequent phase shift acquired by the beam is greatest near the entrance face ($\text{OPL} < z_R$). We assume that the beam is affected only by refractive index changes at $\text{OPL} < z_R$ and that index changes beyond this region are negligible during the period in which diffraction rings are observed (~ 40 s). The interference cones originating from this region therefore diverge *linearly* through the rest of the medium and are projected as rings on the exit plane ($\text{OPL} = 8.82$), which under linear conditions falls in the far field.

To further test the assumption that most of the contribution towards self-phase modulation occurs at $\text{OPL} < z_R$ in the organosiloxane, the experiment was repeated after replacing the sample with $\text{OPL} = 8.82$ mm with one of $\text{OPL} = 0.82$ mm ($\sim z_R$). The spatial intensity profile at the exit face of this sample was projected through air

and onto a plane that was positioned at a distance of 46 mm in the far field. An increasing number of diffraction rings emerged over time with > 13 rings observed at 200 s (Figure 4.3). That a similar maximum in the number of rings was observed in both samples indicates that index changes occurring in the thin sample with $OPL = 0.82$ mm are sufficient to induce a phase shift that is comparable to the one induced in the sample with $OPL = 8.82$ mm. This is consistent with the assumption that self-phase modulation of the beam occurs within $< z_R$ even in the sample with the much longer pathlength. That self-phase modulation is elicited by refractive index changes confined to a narrow region near the entrance face was also postulated in the treatment of nonlinear light propagation in Na vapour [81]. The observation of the conical microstructure in the organosiloxane as well as the emergence of a comparable number of diffraction rings in samples with $OPL < z_R$ and $\gg z_R$ now provide conclusive evidence for this mechanism.

4.4 Temporal evolution of diffraction rings

In most materials employed in previous studies, diffraction rings due to self-phase modulation formed virtually instantaneously and could only be observed in steady state. The emergence of rings in the organosiloxane however could be traced over time (Figure 4.2). This is a direct consequence of the noninstantaneous photoresponse of the medium [27; 106]. The index change due to polymerization at a particular point in time (t) and space as expressed by Equation (4.2) is determined by polymerisation kinetics and it occurs on the order of ms and s. Self-phase modulation therefore takes place on this same timescale and leads to the relatively slow evolution of diffraction rings observed in Figure 4.2: the beam initially diverges from its width of $20\text{ }\mu\text{m}$ at the entrance face to $185\text{ }\mu\text{m}$ at the exit face (Fig 4.2a). Within the next 4 s, the beam develops a single bright ring (Fig. 4.2b) indicating that Δn_0 (and therefore $\Delta\psi_0$) induced during this time is sufficient to induce a maximum phase shift of at least 2π , leading to constructive interference of radiation propagating from the regions about the inflection point, $(d\Delta\psi/dr)_{max}$. As Δn_0 becomes even larger, an increasing number of bright concentric rings due to constructive interference appear in rapid succession (Fig. 4.2 d-g). The number of diffraction rings (N) observed is proportional to the maximum phase shift at a given point in time, as approximated through [6]

$$N \approx \frac{\Delta\psi_0}{2\pi} \quad (4.4)$$

The temporal plot in Figure 4.4a shows the change in the number of diffraction rings over time, which in turn reflects the temporal variation in the maximum phase shift and maximum refractive index change in the organosiloxane (Figure 4.4b). Only 1 ring was experimentally observed in the first 5 s after which there was a rapid increase to 6 rings in the next 10 s. The number of rings observed then increased more gradually over the next 20 s to a maximum of 13 rings. The trend observed in Figure 4.4a is consistent with the rate of refractive index change $\Delta n(r)$ in the medium as shown

in Figure 4.4b, which being directly proportional to $\Delta\psi(r)$, determines the kinetics of self-phase modulation (Equation (4.1)). The gradual saturation of N reflects the exponential decay of $\Delta n(r)$ due to saturation of polymerisation over time (Figure 4.4b).

Figure 4.5a is a temporal plot of the diameter (D) of the outermost ring, which is related to its half-angle β in the following way

$$\beta = \arctan(D/2z) \quad (4.5)$$

where z is the distance from the origin of the interference cones to the plane onto which they are projected. Equation (4.5) assumes that rings propagate linearly after the most significant changes in refractive index had occurred giving rise to self-phase modulation. For the values of diameter of the outermost ring in Figure 4.5a and any value between z_R and 6mm for z , the arctan function remains linear. Therefore the linear increase observed in D corresponds to a linear increase in half-angle (β). Then β can be related to the maximum wavevector $k_{\perp} = (d\Delta\psi/dr)_{max}$ through [6]

$$\beta \simeq \left(\frac{d\Delta\psi}{dr} \right)_{max} / (2\pi/\lambda) \quad (4.6)$$

Based on Equations (4.6) and (4.5) for the half angle (β), the linear increase of the diameter of the outermost ring in Figure 4.5a indicates a linear increase over time in the slope about the inflection point of the phase-shift curve, $(d\Delta\psi(r)/dr)_{max}$, and thereby the slope of the induced refractive index profile, $(d\Delta n(r)/dr)_{max}$. This is illustrated in the scheme of Figure 4.5b, where the transverse phase change has been plotted for induced Gaussian phase shift profiles with increasing central maximum, assuming that $\Delta\psi(r) = \Delta\psi_0 \exp(-2r^2/a^2)$, where $\Delta\psi_0$ is the maximum phase shift, and a is a constant [6]. Here the phase shift profiles with greater $\Delta\psi_0$ present the steeper slopes in the inflexion point or $(d\Delta n(r)/dr)_{max}$. Therefore, the linear increase over time of $(d\Delta\psi(r)/dr)_{max}$ gives an indication that the shape of the refractive index profile induced increases in its maximum value and steepens overtime.

The latter is characteristic of media that undergo intensity-dependent changes of refractive index (e.g. Equation (4.3)). Here, the index profile induced by a Gaussian beam undergoes the greatest rate of change in the most intense, axial region. The resulting steepening of the gradient about the inflection point of the $\Delta n(r)$ curve causes a corresponding strengthening of the $\Delta\psi(r)$ curve and in turn, an increase in β over time.

At later stages, the central regions of the diffraction rings acquire more intensity due to the onset of self-focusing and self-trapping of the beam (Figure 4.2 (f) and (g)). After approximately 100 s, self-focusing is the only dominant process and is evidenced by a significant increase in intensity and complementary narrowing of the beam (Figure 4.2 h) [Chapter 3]. Based on the maximum number of diffraction rings

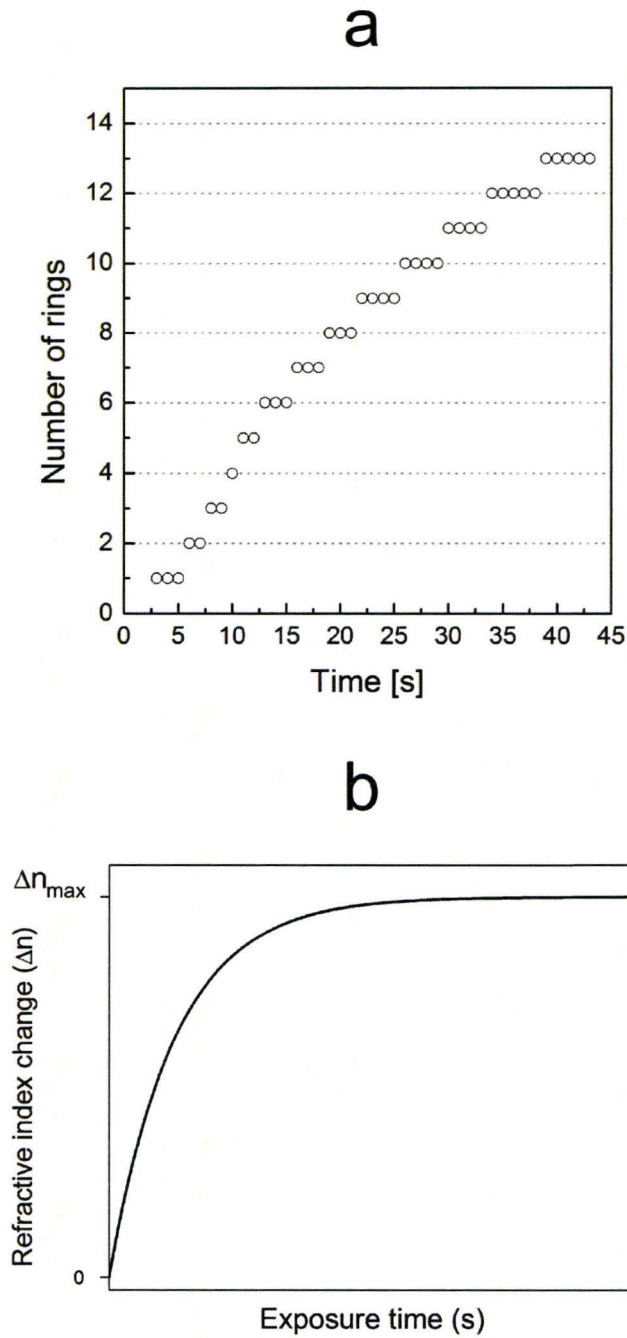


Figure 4.4: (a) Variation of the number (N) of diffraction rings observed at the exit face of the organosiloxane with $OPL = 8.82$ mm as a function of time for an input intensity of 1.6 W/cm^2 . The number of open circles grouped together indicates the number of seconds over which there was no change in number of rings. In (b) a plot of the refractive index change versus exposure time is shown corresponding to Equation (4.2)

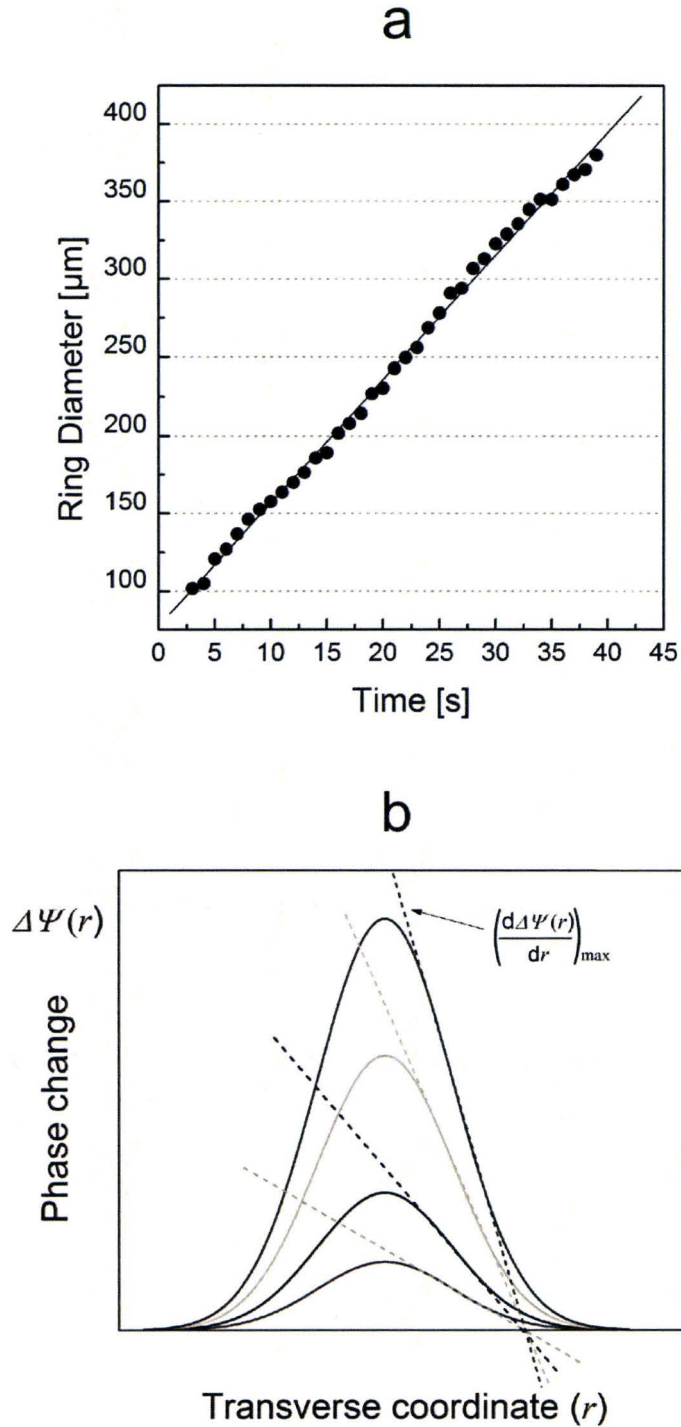


Figure 4.5: (a) Diameter of the outermost ring observed at the exit face of the organosiloxane with $OPL = 8.82$ mm as a function of exposure time. Regression analysis shows a linear relationship between the ring diameter and time with $R^2 = 0.997$. (b) Scheme of phase change profiles for increasing light exposure. Dashed lines show the maximum slope which represent the perpendicular propagation wavevectors $k_{\perp}$.

observed prior to self-focusing ($N = 13$) and assuming that significant refractive index changes occur only in regions within z_R , the maximum Δn_0 achieved immediately prior to the onset of self-focusing was calculated to be 0.008, which is close to the maximum change in refractive index ($\Delta n_s = 0.006$) induced in organosiloxane films [24]. This indicates that once saturation of the refractive index is achieved in regions within z_R , index changes beyond this region become significant enough to counter the natural diffraction of the beam, enabling it to self-focus.

4.5 Intensity dependence of self-phase modulation

As mentioned earlier, self-phase modulation in the organosiloxane could be induced only within a relatively narrow range of intensities (1.6 W/cm^2 to 16 W/cm^2). We previously found that the profile of $\Delta n(r)$ varies significantly with intensity and in turn, determines the type of nonlinear event that is elicited in the medium. In the high intensity regime ($\geq 159 \text{ W/cm}^2$), the refractive index rapidly saturates over a large transverse area of the beam; the Gaussian profile in the $\Delta n(r)$ curve required for self-phase modulation cannot be achieved and the beam suffers filamentation (Chapter 3 and [21]). By contrast, index changes induced in the low intensity regime ($\leq 0.16 \text{ W/cm}^2$) are insufficient to impose phase shifts $> 2\pi$ and diffraction rings cannot be elicited under these conditions. Instead the beam self-focuses and self-traps over a time-scale of $\approx 30 \text{ s}$ due to refractive index changes along its propagation axis. The average Δn required for self-trapping can be estimated by calculating the refractive index of a single-mode, $48 \text{ }\mu\text{m}$ -wide, gradient-index cylindrical waveguide at 532 nm . The resulting value of 1.3×10^{-5} is at least an order of magnitude smaller than the index change of $\approx 6 \times 10^{-4}$ required to generate 1 diffraction ring through self-phase modulation. Collectively, the observations at the high and low intensity regimes suggest that a balance is struck in the mid intensity regime (1.6 W/cm^2 - 16 W/cm^2) within which both the magnitude and profile of the induced refractive index satisfy the conditions required for self-phase modulation. Moreover, this $\Delta n(r)$ profile is induced within a timescale ($\approx 4 \text{ s}$) at which index changes in regions $> z_R$ - and therefore self-trapping - can be considered negligible.

4.6 Curvature dependence

In the experiments described in the previous Sections, diffraction rings were induced by a Gaussian beam with its waist incident at the entrance face of the sample. The radius of curvature of the beam in this plane is therefore ∞ . In this Section, we show that entirely different types of rings can be induced by varying the curvature of the beam that is incident on the sample.

Previous studies have shown that the wavefront curvature (R) of the incident laser beam influence resulting patterns of diffraction rings (Section 1.3.3). For example,

experimental studies on liquid crystal films showed that diffraction rings with a bright central spot were induced in the far-field by a divergent beam ($R > 0$) whereas rings with a dark central spot were induced by a convergent beam ($R < 0$) [82]. The latter has also been observed in defocusing media due to thermally induced refractive index changes, such as chromophore-doped liquids when $R > 0$ [84].

Experiments of the dependence of wavefront curvature have until now only been performed in thin samples (pathlengths $< z_R$) where diffraction rings were observed in the far-field. As detailed in the following Sections, we have carried out detailed studies including statistics of the effect of beam curvature in the induced diffraction rings and the temporal evolution and dynamics of diffraction rings propagating over long distances ($z \gg z_R$) in the organosiloxane.

4.6.1 Experimental configuration

Figure 4.6 is a scheme of the experimental configuration for experiments of curvature dependence. The intensity at the beam waist was 1.6 W/cm^2 ($5 \mu\text{W}$ of average power) and its diameter ($2\omega_0$), $20 \mu\text{m}$. The beam was launched in a sample with a pathlength of 6 mm (OPL= 8.82 mm). The entrance face of the sample was placed to the left (2 mm) of the beamwaist for a negative input wavefront curvature, $R < 0$, (Figure 4.6b) and to the right (2 mm) of the beamwaist for a positive curvature, $R > 0$ (Figure 4.6c). The error in positioning the sample was approximately $\pm 0.25 \text{ mm}$. Further details of the experimental configuration are provided in Section 2.3.

4.6.2 Effect of $R < 0$ and $R > 0$ in organosiloxane

The mechanism of self-phase modulation as expressed by Equation (4.1) does not take into account the transverse phase shift originating from wavefront curvature. In order to do so, the complex amplitude of a TEM_{00} Gaussian beam launched at the entrance of the sample must be considered [85]

$$E(r, z_0) = E(0, z_0) \exp\left(-\frac{r^2}{\omega^2}\right) \exp\left(-\frac{ik_0 n_0 r^2}{2R}\right) \quad (4.7)$$

where k_0 and n_0 are the wavenumber and refractive index in free-space, respectively, r is the radial coordinate, z_0 is the position of the entrance face of the medium and R is the radius of wavefront curvature. Then the *total* transverse phase shift induced by the beam at a distance d from the input plane of the medium becomes [85]

$$\psi(r) = \frac{k_0 n_0 r^2}{2R} + \Delta\psi(r) \quad (4.8)$$

Substituting Equation (4.1) for $\Delta\psi(r)$ [85],

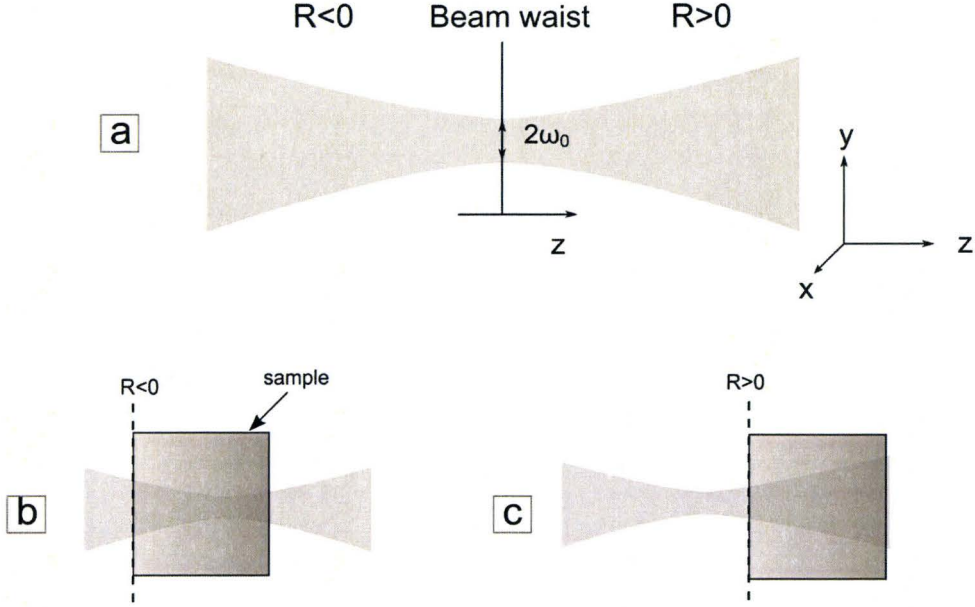


Figure 4.6: Scheme of a) the beamwaist and regions of positive and negative curvatures (R) and examples of configurations for b) convergent ($R < 0$) and c) divergent ($R > 0$) beams incident on the input face of a sample. The beam propagates in the positive z direction.

$$\psi(r) = \frac{k_0 n_0 r^2}{2R} + \int_{z_0}^{z_0+d} \Delta n(r, z) dz \quad (4.9)$$

where the first and second terms correspond to the contributions of the curvature (R) and the change in refractive index, respectively, to the total transverse phase shift. The resulting diffraction ring patterns will depend on the sign of the *product* of the first and second terms of Equation (4.8) [85].

As shown below, diffraction rings with a dark centre are induced when $R < 0$ whereas rings with a bright centre are obtained when $R > 0$.

The first term in Equation (4.8) is the contribution of wavefront curvature to the phase shift. For the contribution from changes in refractive index we assume a Gaussian shape using[6]

$$\Delta\psi(r) = \Delta\psi_0 \exp(-2r^2/a^2) \quad (4.10)$$

where a is the beam radius. Because the organosiloxane is a self-focusing medium that undergoes positive changes in refractive index, $\Delta\psi(r) > 0$. The sum of both contributions, gives the total phase shift which is different for positive and negative curvatures (Figure 4.7).

The variation of the total phase shift at different positive and negative curvatures can be seen in Figure 4.8a and 4.8b, respectively. The greater the value of $|R|$,

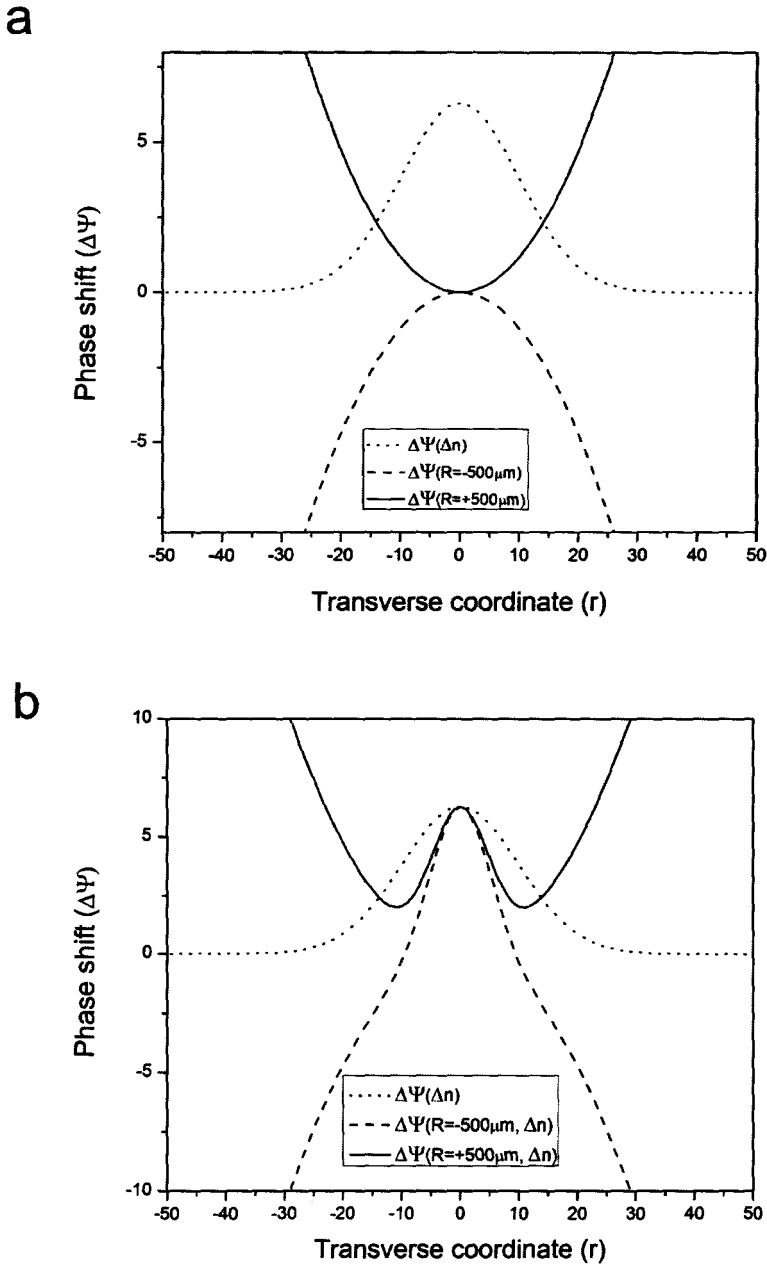
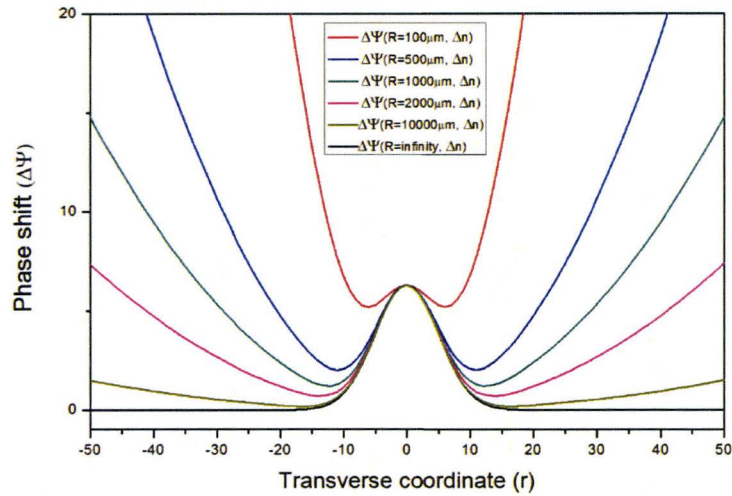


Figure 4.7: a) Contributions of Δn and positive and negative curvatures to the total phase shift $\Delta\psi$ and b) total contribution of Δn plus positive and negative curvatures to $\Delta\psi$. In b) contribution of Δn is only shown for comparison.

a) Total phase shift for $R > 0$



b) Total phase shift for $R < 0$

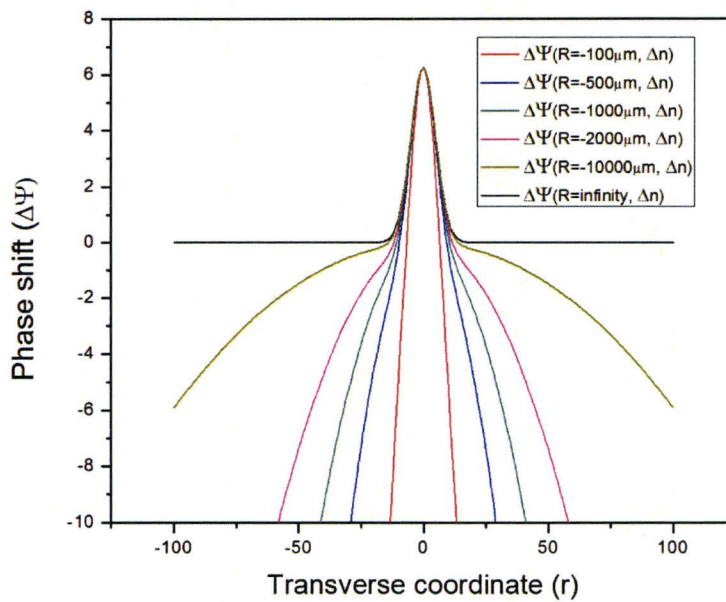


Figure 4.8: Total phase shift for different contributions of a) positive and b) negative curvatures.

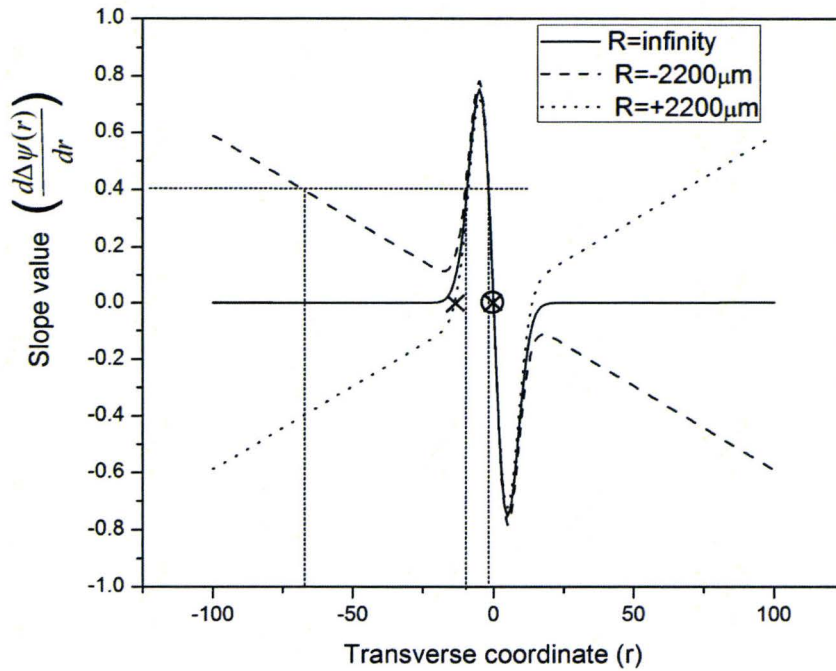


Figure 4.9: Derivatives of total phase shifts for a fixed Δn contribution and contributions of a positive curvature, $R=+2200 \mu\text{m}$ and a negative curvature $R=-2200 \mu\text{m}$. The derivative for the contribution of an infinite curvature is shown for comparison.

the smaller the difference in total phase shift profile relative to that when $R = \infty$. The slope of the total phase shift curves at a particular point r corresponds to the perpendicular wave vector $\kappa_{\perp} = d\Delta\psi/dr$. The curves corresponding to the derivative of the total phase shift for a positive ($R=+2200 \mu\text{m}$) and a negative curvature ($R=-2200 \mu\text{m}$) are shown in Figure 4.9. Whenever the same value of wave vector (or slope) exists for two points in r , based on $\Delta\psi(r_1) - \Delta\psi(r_2) = m\pi$, constructive (m even) or destructive (m odd) interference can take place. For example, the dotted line in Figure 4.9, shows that two r points and three r points share the slope value of 0.4, for positive and negative curvatures, respectively. Clearly interference occurs in both cases and interference occurs mainly between the two r points where most of the energy concentrates. Analyzing only half of the profile, since it is symmetric along $r = 0$, for the negative curvature, the values of slope that are not shared by two points in r are around zero, this is indicated by a red circle in Figure 4.9. This means that around those values no interference occurs. These values correspond to the slopes of the central part of the beam and points nearby, therefore this explains the dark zones formed at the center surrounded by rings for negative curvatures. For a positive curvature, values of slope of zero and around this value are held by two points in r , which is marked with the blue crosses in Figure 4.9. This could explain the observation of a bright spot in the center of the rings for positive curvatures. This is consistent with the patterns predicted by Deng and coworkers [85] where rings with a dark central spot are observed for $R < 0$ and rings with a bright central spot are observed for $R > 0$ in a self-focusing medium.

Figure 4.10 shows the total phase shift for positive and negative curvatures for $|R|=2200 \mu\text{m}$ with varying Δn .

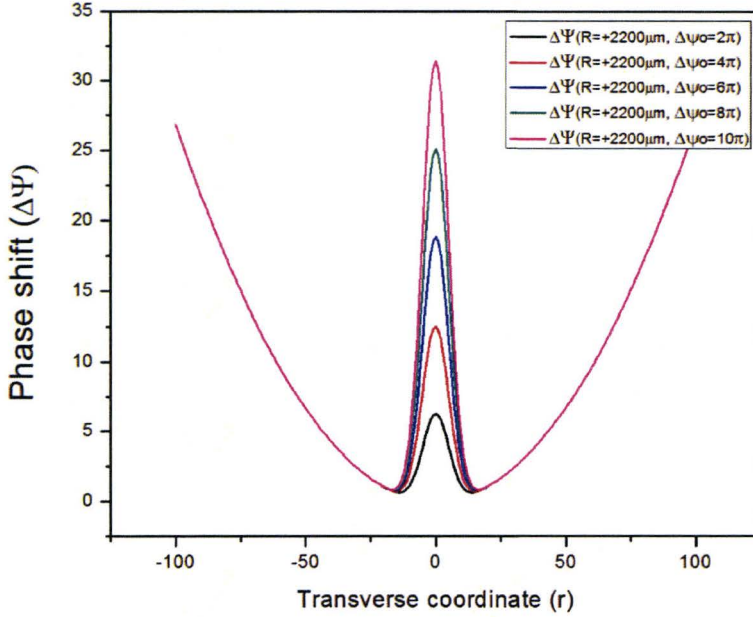
To do that, we varied $\Delta\psi_0$ in Equation (4.10) from 2π to 10π . The shape of the total induced phase shift remains the same but an increase in the maximum phase shift is observed for both positive and negative curvatures as the Δn contribution is increased. When plotting the derivatives of the total phase shifts in Figure 4.11, we see that an increase in the absolute value of the slope occurs when increasing the Δn contribution for both positive and negative curvatures. As expected from theory, the greater the refractive index change, the greater the maximum phase shift and the more slope values exist that are shared by two r points. Therefore more wavefronts can interfere and a greater number of diffraction rings can be induced.

4.6.3 Statistics of results for $R < 0$ and $R > 0$

Figure 4.12 shows the 2D and 1D intensity profiles of diffraction rings with a dark and bright centre induced after propagation of beams with negative ($R < 0$) and positive curvature ($R > 0$), respectively.

Table 4.1 shows statistics of experimental results obtained at positive and negative input beam curvatures in the organosiloxane. There was excellent agreement with

a) Total phase shift for $R>0$ and increasing Δn



b) Total phase shift for $R<0$ and increasing Δn

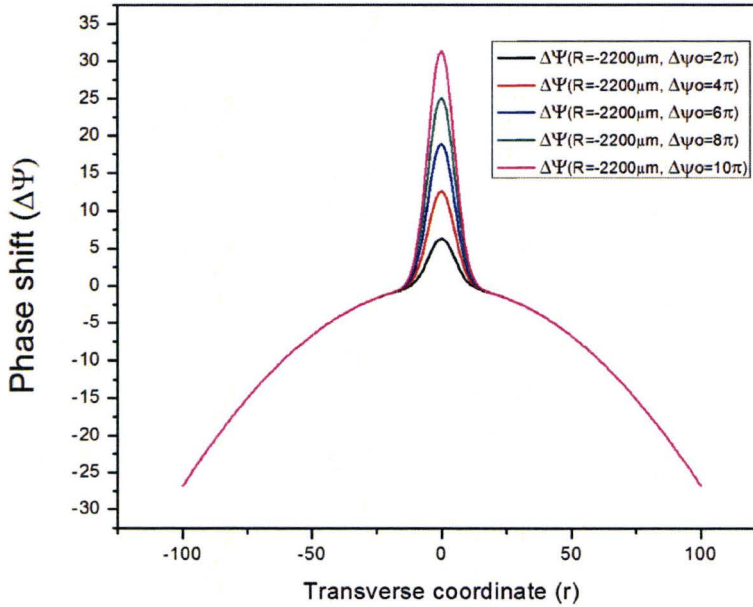
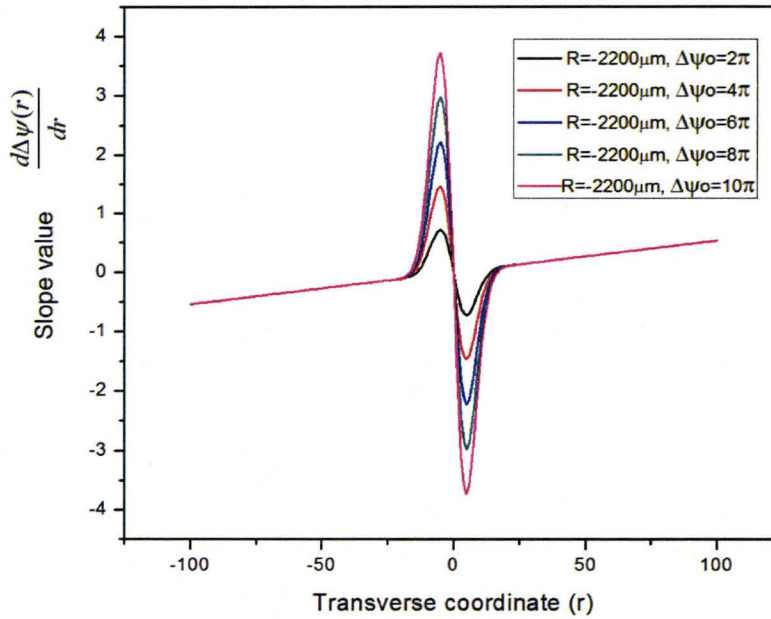


Figure 4.10: Total phase shift for a) positive and b) negative curvatures with different contributions of Δn by increasing $\Delta\psi_0$.

a) $R > 0$ and increasing Δn



b) $R < 0$ and increasing Δn

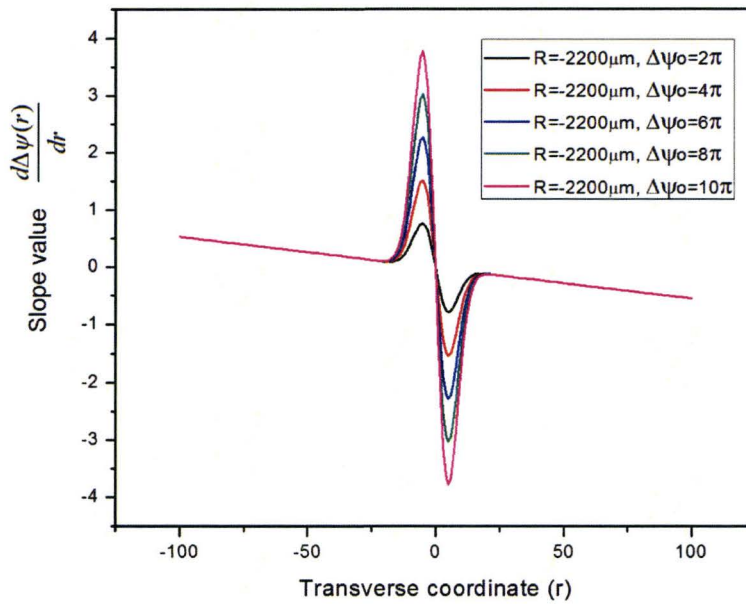


Figure 4.11: Derivatives of total phase shift for a) positive and b) negative curvatures with different contributions of Δn by increasing $\Delta\psi_0$.

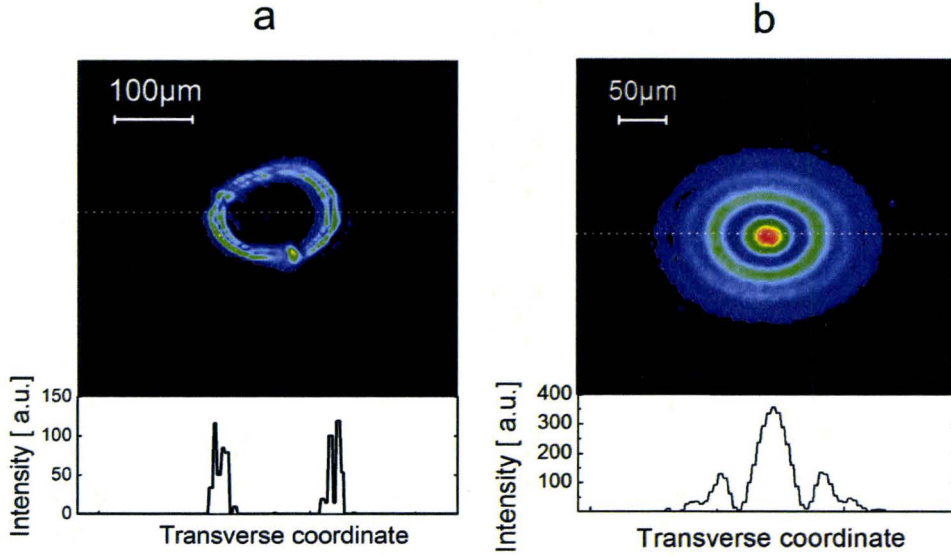


Figure 4.12: Diffraction rings for a) $R < 0$ with a dark central spot and b) for $R > 0$ with a bright central spot. 2D and 1D intensity profiles are shown.

Table 4.1: Statistics of beam curvature dependence on ring patterns

Beam curvature	Radius of curvature (mm)	Distance from beam waist	Observed ring patterns			# experiments
			Bright spot	Big Dark spot	Fingerprint	
$R < 0$	$R = -2.2$	2 mm		100%		12
$R > 0$	$R = +2.2$	2 mm	62%	31%	12%	26

theory for experiments carried out at $R < 0$. For $R = -2.2 \text{ mm}^\dagger$, 100% of experiments yielded diffraction rings with a dark centre. Qualitatively, this ring pattern is similar to the one induced at $R = \infty$ (Section 4.4). However, the dark central spot for rings induced at $R = -2.2 \text{ mm}$ was much bigger. As detailed in Section 4.6.4 the dynamics and temporal evolution of the rings were also significantly different.

The majority of experiments (62%) carried out at $R > 0$ yielded diffraction rings with a bright central spot, which is consistent with theory. In 31% of cases, however, rings with a dark centre were obtained. These showed similar behaviour to those induced at $R = -2.2 \text{ mm}$. This observation could be explained by the complex dynamics along the pathlength. Theory assumes that the contribution of the radius of curvature R to the total phase shift occurs in a thin medium, however in our case a thick medium is employed. During the propagation of the beam, under certain conditions of pre-polymerization of the sample, the beam could begin to self-focus acquiring a wavefront with $R < 0$ and therefore obtaining rings with a dark centre at the output face. In the case of experiments with an initial beam with $R < 0$, even

[†]The radius of curvature was calculated using the formula $R(z) = z[1 + (\pi\omega_0^2/\lambda z)^2]$ for a Gaussian beam [34].

if the beam experiences self-focusing, the curvature remains negative and we always observe rings with a dark centre. Another type of rings that we named *fingerprint* rings, were observed in 12% of experiments. Details of this type of rings will be discussed in Section 4.7.

4.6.4 Dynamics of rings with dark central spot for $R < 0$

The temporal evolution for a beam with initial curvature of $R = -2.2$ mm (2 mm to the left of the beam waist) is shown in Figure 4.13. Initially, the convergent Gaussian beam propagates from an approximate radius of $35\text{ }\mu\text{m}$ [†] at the entrance face of the organosiloxane to $36\text{ }\mu\text{m}$ at the output face (Figure 4.13a). The apparent lack of diffraction is due to the convergence of the beam at the entrance face of the sample (Figure 4.6).

Over time refractive index changes induce spatial self-phase modulation, which leads to the emergence of a single ring at 7 s (Figure 4.13b) with a dark centre with 20 a.u.. This corresponds to 9 % of the maximum intensity along the ring. This value agrees with intensities of central dark spots in previous theoretical and experimental studies of self-phase modulation in thin films [85; 84]. The central dark spot increases in diameter over time (Figures 4.13b-f) and darkens until reaching 0 a.u. of intensity. At the same time, the ring surrounding the dark spot increases in diameter from approximately $70\text{ }\mu\text{m}$ to $210\text{ }\mu\text{m}$ and multiple rings (up to 4) emerge around the dark central spot (Figure 4.13g). At 71 s, each of the diffraction rings started to spontaneously break up into a discrete series of filaments, that were positioned with azimuthal symmetry. Filamentation continued up to 131 s (Figure 4.13j), at which point, approximately 13 individual filaments had formed. Filaments observed in the innermost ring contained the greatest intensity and radial traces towards the outward rings were observed. Filamentation was accompanied by the onset of self-focusing, which was observed as an increase in intensity in the centre of the rings. Self-focusing was accompanied by a decrease in intensity from the surrounding filaments and rings. At 198 s, the self-trapped beam was the dominant feature with a relative intensity of 700 a.u.

In the organosiloxane, we observed an increase in dark spot diameter over time as seen in Figure 4.14. The change of dark central spot diameter as a function of exposure time shows that it roughly follows the kinetics of photopolymerization, with a sharp increase initially and a saturation of the diameter overtime. To explain this behaviour, we can examine the phase shift induced over time and its derivatives. When the refractive index increases, its contribution to the total phase shift increases as seen in Figure 4.10b for $R < 0$. In order to observe an increase in the central dark spot diameter, we would expect that in the derivatives of the total phase shifts, an

[†]The beam radius at a particular z was calculated using the formula $\omega(z) = \omega_0[1 + (\lambda z/\pi\omega_0^2)]^{1/2}$ for a Gaussian beam [34].

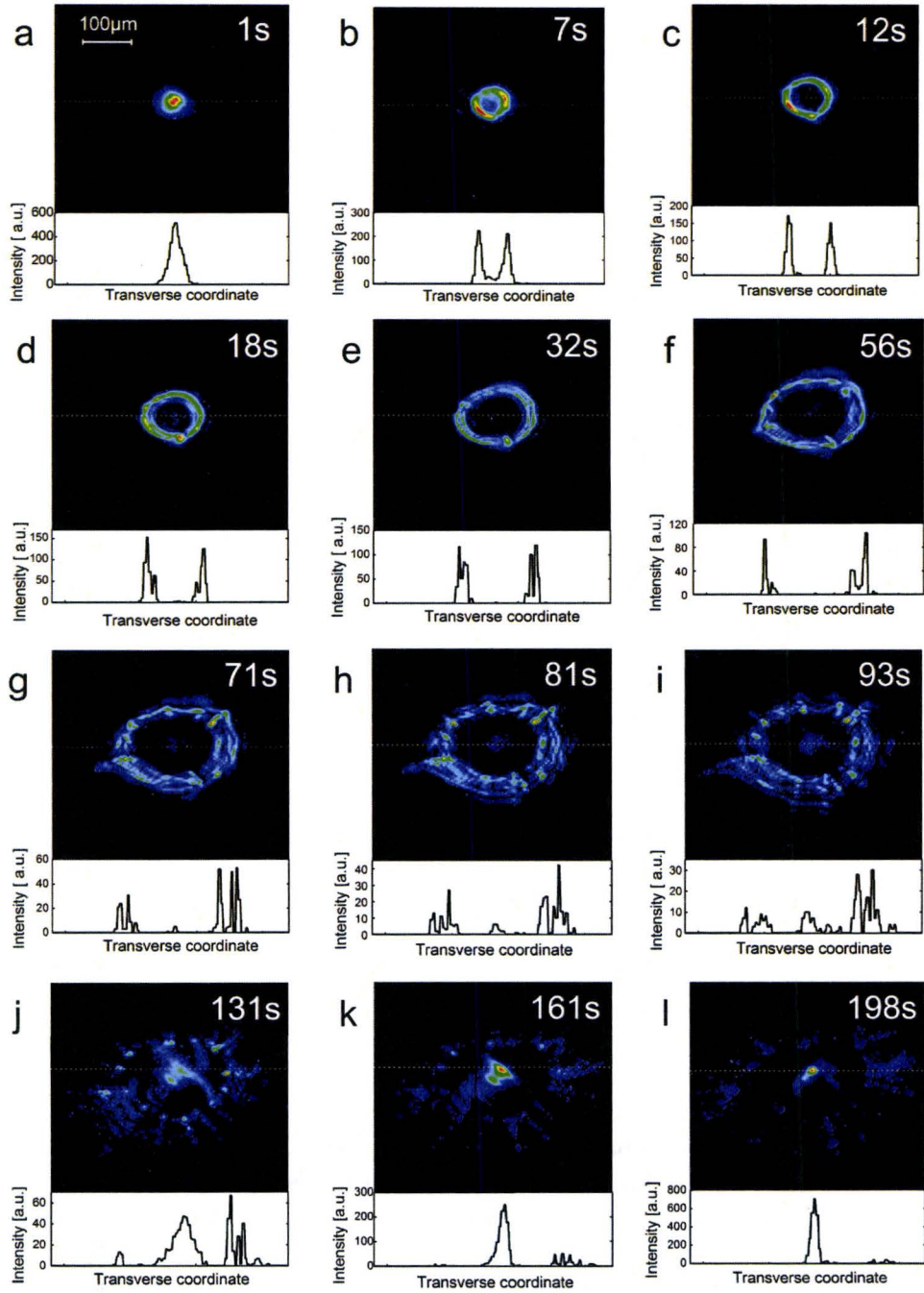


Figure 4.13: Temporal evolution of rings with dark central spot for an input beam with curvature of $R = -2.2$ mm at 1.6 W/cm². 2-D and 1-D spatial intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value.

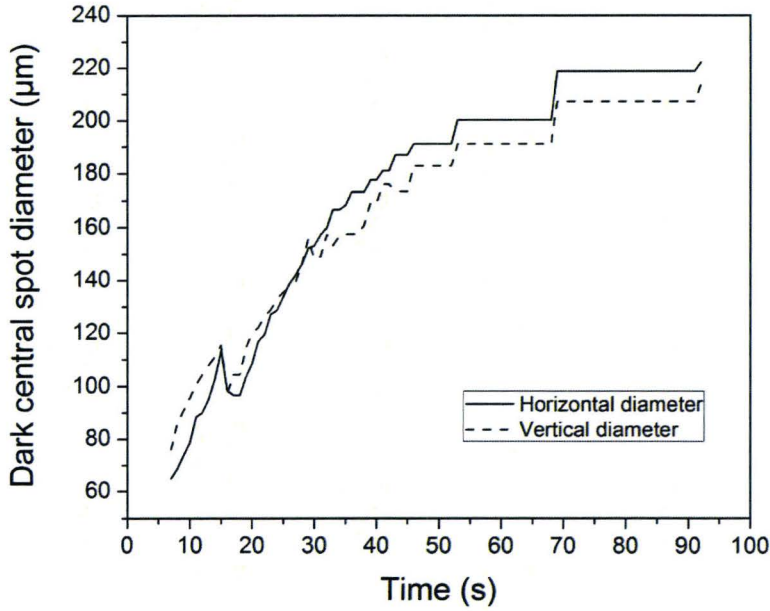


Figure 4.14: Diameter of the dark central spot observed at the exit face of the organosiloxane with $OPL = 8.82$ mm as a function of exposure time for an input beam with curvature of $R = -2.2$ mm at 1.6 W/cm².

increasing number of r values around $r=0$ would not share the slope value with any other r point as the refractive index increases. And that way no interference would occur over a larger area around $r=0$. However, when examining the derivatives in Figure 4.11b, there is no indication of such behaviour.

We realize that over time, as exposure increases, areas that did not initially contribute significantly to refractive index changes now do so. This implies an increase in effective beam diameter for contributing to refractive index changes and thereby, the total phase shift. When plotting the derivatives of the total phase shift for an increasing beam radius (Figure 4.15a) and fixed Δn contribution, we see the expected behaviour of increasing number of r values around $r=0$ that do not share a slope value. This indicates an increase in the dark central spot diameter. However, we observe that fewer r points share the same slope value also away from $r = 0$, implying that fewer diffraction rings are induced, which does not agree with the experimental observations. If the Δn is increased along with the beam radius as shown in Figure 4.15b, then the derivatives of the total phase shift show both the increase of the central dark spot diameter and the existence of multiple diffraction rings over longer exposure.

The increase in diameter of the dark central spot has also been observed in absorbing liquid media that undergo thermally induced refractive index changes, such as Rhodamine 6G diluted with methanol [84]. In these experiments, a divergent input beam was introduced into a self-defocusing medium. This yielded similar ring

$R < 0$

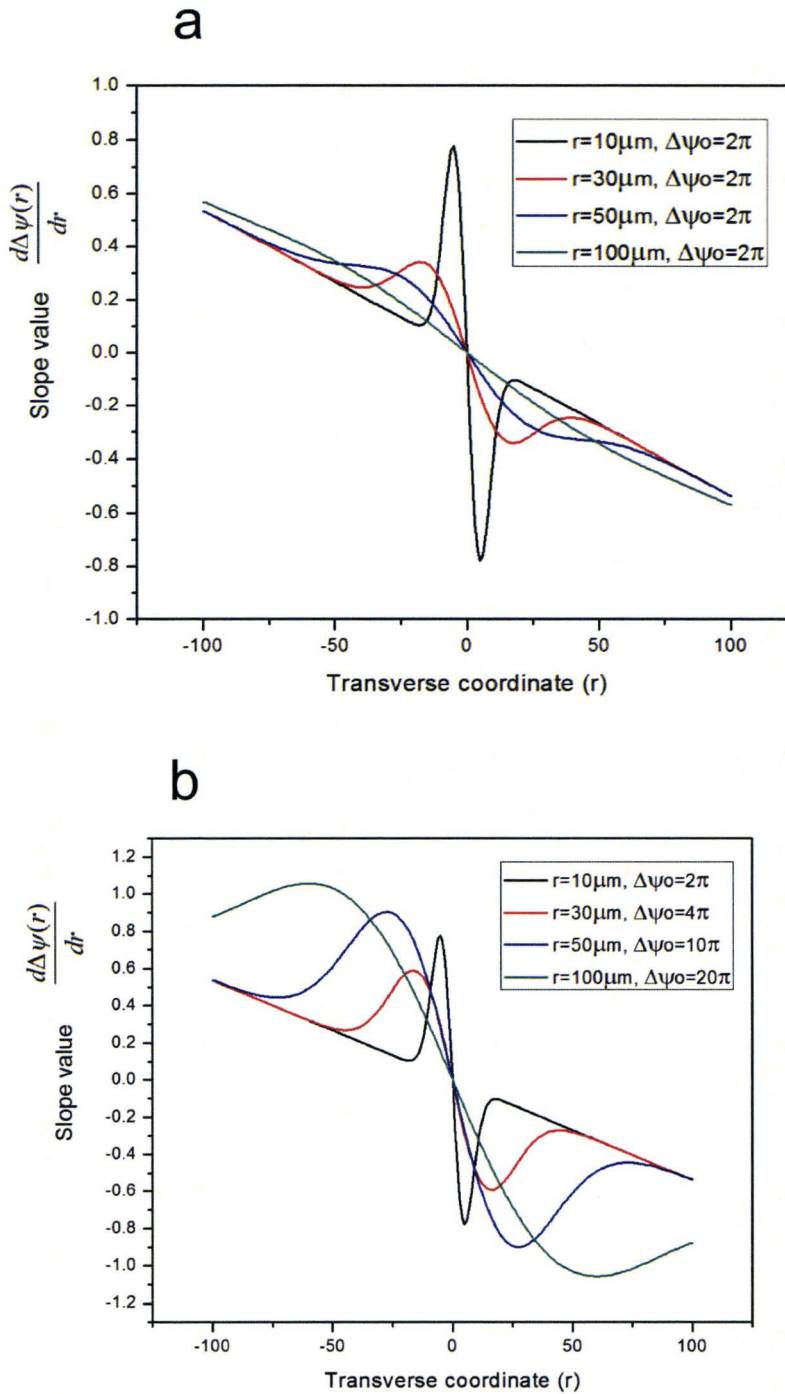


Figure 4.15: Derivatives of total phase shift for $R = -2.2 \text{ mm}$ with a) varying beam radius and b) varying beam radius and Δn contribution.

patterns as the ones we obtained with a convergent beam and self-focusing medium (see Section 1.3.3). However, the dark spot diameter in this study increased linearly with pump power whereas in our experiments, it increased over time at a single input power.

Filamentation of diffraction rings occurring at later stages in our experiment, has not been observed in other studies of the effect of beam curvature on self-phase modulation and was not observed in the organosiloxane with input beams at the beam waist ($R = \infty$).

Filamentation can be explained by modulation instability theory, which states that amplitude and phase perturbations (noise) in plane waves can develop and grow exponentially over the propagation path, until their strength is comparable to the initial plane-wave field, ultimately breaking up into multiple beams or filaments [96]. In our group, modulation instability in the organosiloxane leading to multiple filaments has been previously shown [106].

Filamentation of diffraction rings forming at $R = -2.2$ mm occurs due to the timescale of events. As seen in Figure 4.13, self-trapping at $R = -2.2$ mm occurred until 198s, much later compared to experiments at $R = \infty$, which occurred at approximately half of the time (Figure 4.2h). This coupled with the fact that diffraction rings around the dark spot do not significantly change in number or size from 56s to 131s (Figures 4.13f-j), gives sufficient time for the beam to be susceptible to transverse noise resulting in instability of the diffraction rings and their rupture into individual filaments. The filaments at 93 s (Figure 4.16), were positioned with azimuthal symmetry. This originates from the non-radially symmetric noise being imposed on the diffraction rings, however the noise could possess either linear symmetry or non-azimuthal symmetry as filaments along the rings are aligned with respect to each other. For diffraction rings induced with positive curvatures $R > 0$ (see next Section) dynamics of the rings induces continuous changes to the rings profiles, therefore filamentation does not occur.

4.6.5 Dynamics of rings with a bright central spot for $R > 0$

The temporal evolution of rings emerging with a bright central spot for an input beam curvature of $R = +2.2$ mm (2 mm to the right of the beamwaist) is shown in Figure 4.17. The Gaussian beam initially diffracts from a radius of approximately $35\text{ }\mu\text{m}$ to $206\text{ }\mu\text{m}$ (Figure 4.17a). At 15 s, the first ring develops around a bright central spot (Figure 4.17b). This is consistent with the predicted pattern of diffraction rings induced at positive curvatures in self-focusing media [85]. In the next 12 s, increasing number of bright concentric rings appeared with a maximum of 5 rings at 27s (Figures 4.17c-e). It is likely that more rings emerged but they might not be visible due to the high intensity of the central spot.

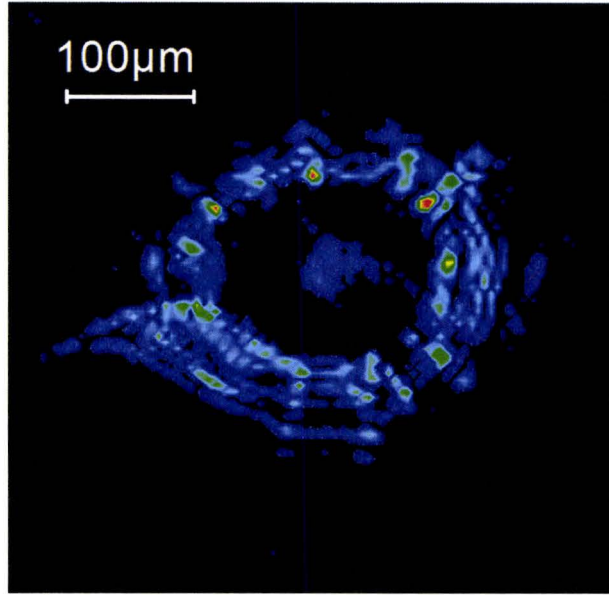


Figure 4.16: Magnified 2D profile of Figure 4.13i showing filamentation of diffraction rings at 93 s for an input beam with curvature of $R = -2.2$ mm at 1.6 W/cm².

With time, exchange of intensity between the bright central spot and the two innermost rings was observed. For example, at 19 s (Figure 4.17c) the bright central spot was greater in intensity with 350 a.u., with the first and second ring at approximately 125 a.u. and 50 a.u., respectively. In the next few seconds, the second ring gained energy and at 27 s, (Figure 4.17e) its intensity was greater than the first ring with 100 a.u. and the bright central spot had decreased in intensity to around 170 a.u. Between 27 s and 36 s the exchange in intensity seemed to occur only between the two innermost rings. At 33 s, the intensity transferred from the second ring to the first and the converse occurred at 36 s. At this point, both rings had a thickness of approximately $25\text{ }\mu\text{m}$. This changed at 40 s, when the intensity concentrated mainly on the bright central spot, reaching 200 a.u. (Figure 4.17h) with the first two rings apparently fused into a thicker ring of $\sim 60\text{ }\mu\text{m}$ of radius and with 70 a.u. This exchange of intensity continued over time and the rings remained for as long the experiment was recorded (926 s) (Figure 4.17).

In theoretical and experimental studies, there is an association of the type of rings with beam curvature and number of rings with maximum phase shift $\Delta\psi_0$ [85; 83]. We confirmed this for positive (and negative) curvatures since we always observed the bright (dark) central spot, and overtime the number of rings increased which was an indication of increase in Δn and therefore $\Delta\psi_0$. This is explained for $R > 0$ with the plots of the derivatives of the total phase shift in Figure 4.11a. The increasing contribution of Δn occurring over time generates more slope values shared by two

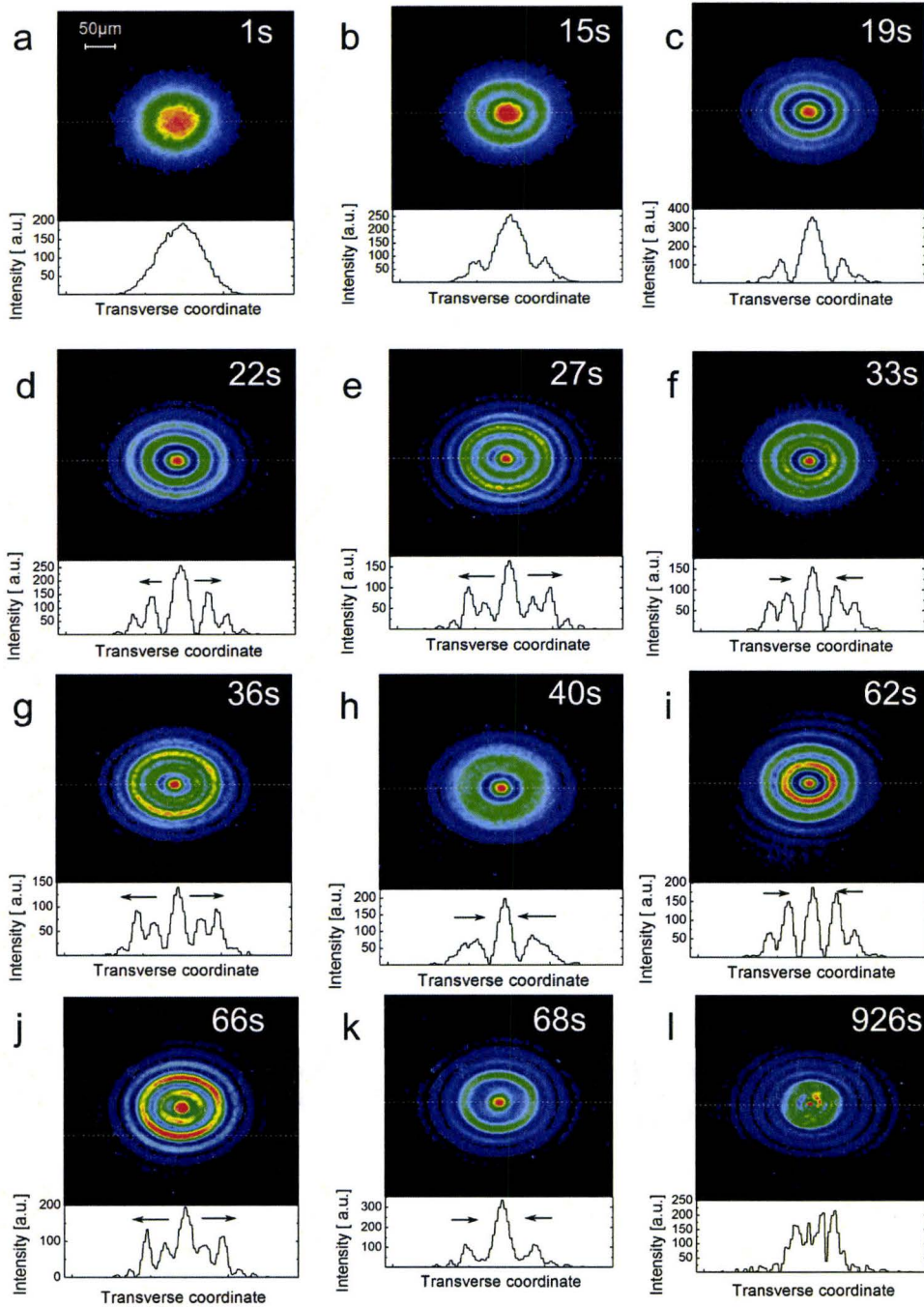


Figure 4.17: Temporal evolution of rings with bright central spot for an input beam with curvature of $R = +2.2$ mm at 1.6 W/cm². 2-D and 1-D spatial intensity profiles of the beam are shown. Arrows indicate the direction of flow in intensity. For clarity, each 2-D profile has been normalized to the maximum intensity value.

points of r which can produce interference and increases the number of diffraction rings induced.

The observed phenomenon of exchange in intensity between the bright spot and the inner rings over time, has not been reported before for other media. This could be explained as all curvature studies have been performed in thin media. The bright rings in our experiments propagate over long distances and therefore as diffraction rings with a bright centre are being induced, complex dynamics involving changes in refractive index and saturation effects occur along the propagation path. Further studies should be performed to understand this process in detail.

A similar phenomenon was observed in a thin self-defocusing Kerr media with negative beam curvatures $R < 0$ [83]. This configuration is equivalent to the self-focusing organosiloxane with $R > 0$ (see Section 1.3.3). In their experiments, as they varied the input power, the distribution of energy and ring configuration changed. For low powers, they observe a set of thin rings uniformly spaced surrounding the central spot and at high powers they saw thicker and brighter rings modulating the thin ones. However, no further details on the mechanism of this phenomenon was given.

4.7 Different types of ring patterns at $R = \infty$

The effect on wavefront curvature on the type of diffraction rings and the dynamics of self-phase modulation was described in the previous Section (4.6). Here we return to self-phase modulation induced at $R = \infty$ and present the emergence of different ring patterns.

Self-phase modulation strongly depends on the refractive index change experienced by the organosiloxane, which determines the induced phase shift and ultimately the emergence of diffraction rings. Even under apparently identical conditions, dynamics within the medium can be affected by minor variations in refractive index giving rise to strikingly different results. Our samples undergo pre-polymerisation prior to experiments of nonlinear light propagation. This process can lead to minor variations in the maximum refractive index change Δn_s for different samples.

At $R = \infty$ the beam waist was incident on the entrance face of the sample and the intensity of the beam was 1.6 W/cm^2 . Sample thicknesses of 0.56 mm, 2 mm, 4 mm, 6 mm, 8 mm and 10 mm corresponding to optical path lengths of 0.82 mm, 2.94 mm, 5.88 mm, 8.82 mm, 11.76 mm and 10.47 mm, respectively were examined. Four types of diffraction patterns were observed under these conditions: rings with a dark centre (dark rings), rings with a bright centre (bright rings), rings with high-order modes and rings that appear all at once (fingerprint rings).

The results presented in this Section were obtained after a careful analysis of the statistics of experimental results at $R = \infty$, which has not been done before in any

media. In our results, in the majority of cases the theoretically expected dark rings were observed, more than 60% for all pathlengths > 2 mm imaged at the exit face or further away (see Figure 4.18). However, we also observed rings with high-order modes (up to 15%) and fingerprint rings (up to 14 %) in a significant number of experiments. These type of rings are likely due to variations in prepolymerisation; high-order modes occurring when Δn_s is relatively large and fingerprint rings forming when Δn_s is relatively small. Error in positioning leads to the observation of bright rings (up to 25%) in some of the experiments. For the shortest pathlength of 0.56 mm bright rings were observed most of the time (94%). For the 2 mm pathlength, imaged at the exit face, an expansion of the beam was mostly (73 %) observed. This could correspond to the near-field diffraction (see Section 4.8). At the far-field, most of the times (71%) diffraction rings of various types are observed.

Dark rings: This type of rings was previously presented in Sections 4.2 to 4.4. It is the type expected theoretically for $R = \infty$ and also the most similar to the one observed in previous studies at the beam waist for other nonlinear media [6; 4]. Dark rings as seen in Figure 4.2, evolve from one to multiple rings over time and ultimately self-focusing of the beam occurs. After the appearance of the first few rings, this pattern appears very similar to rings with a big dark central spot for $R < 0$, however when multiple rings emerge (Figure 4.2 e-g), they appear close to the central zone which is not observed for $R < 0$.

According to Equation (4.8), contributions to self-phase modulation originate only from changes in refractive index ($\int_{z_0}^{z_0+d} \Delta n(r, z) dz$) due to free-radical polymerization. If the phase shift profile follows the initial Gaussian beam shape, then for values of $\Delta\psi_0 \gg 2\pi$, we would expect continuous concentric rings. From Section 4.6.2, the derivative of the total phase shift for $R = \infty$ is more similar to the derivative with negative curvature contributions, in that for slope values around zero, no interference occurs since no two points in r share the same slope value as shown in Figure 4.9. Therefore we can expect to have in the central part of the diffraction ring pattern more concentric rings for $R = \infty$ compare to lower negative R values and perhaps a small dark zone for $R = \infty$ corresponding to the slope value of zero that does not undergo interference. Based on this, the dark ring pattern is the one we would expect to occur at $R = \infty$. Moreover, statistics shown in Figure 4.18a, confirm that these are the most observed rings for OPL 5.88 mm [77 %], 8.82 mm [76 %], 11.76 mm [71 %] and 14.7 mm [64 %], which is consistent with theory.

In previous studies, carried out in nematic liquid crystals [6] and absorbing solutions with thermal dependent index changes [4], the beam waist was also placed at the entrance of the nonlinear medium ($R = \infty$). The observed pattern for high powers or induced values of $\Delta\psi_0 \gg 2\pi$ was dark ring with multiple concentric diffraction rings going from the center outwards, without the presence of a big dark spot, which is consistent with our observations.

Rings with high-order modes: Rings superposed with high-order modes were also observed. After the beam diffracts in the first second to a diameter of $414 \mu\text{m}$ (Figure

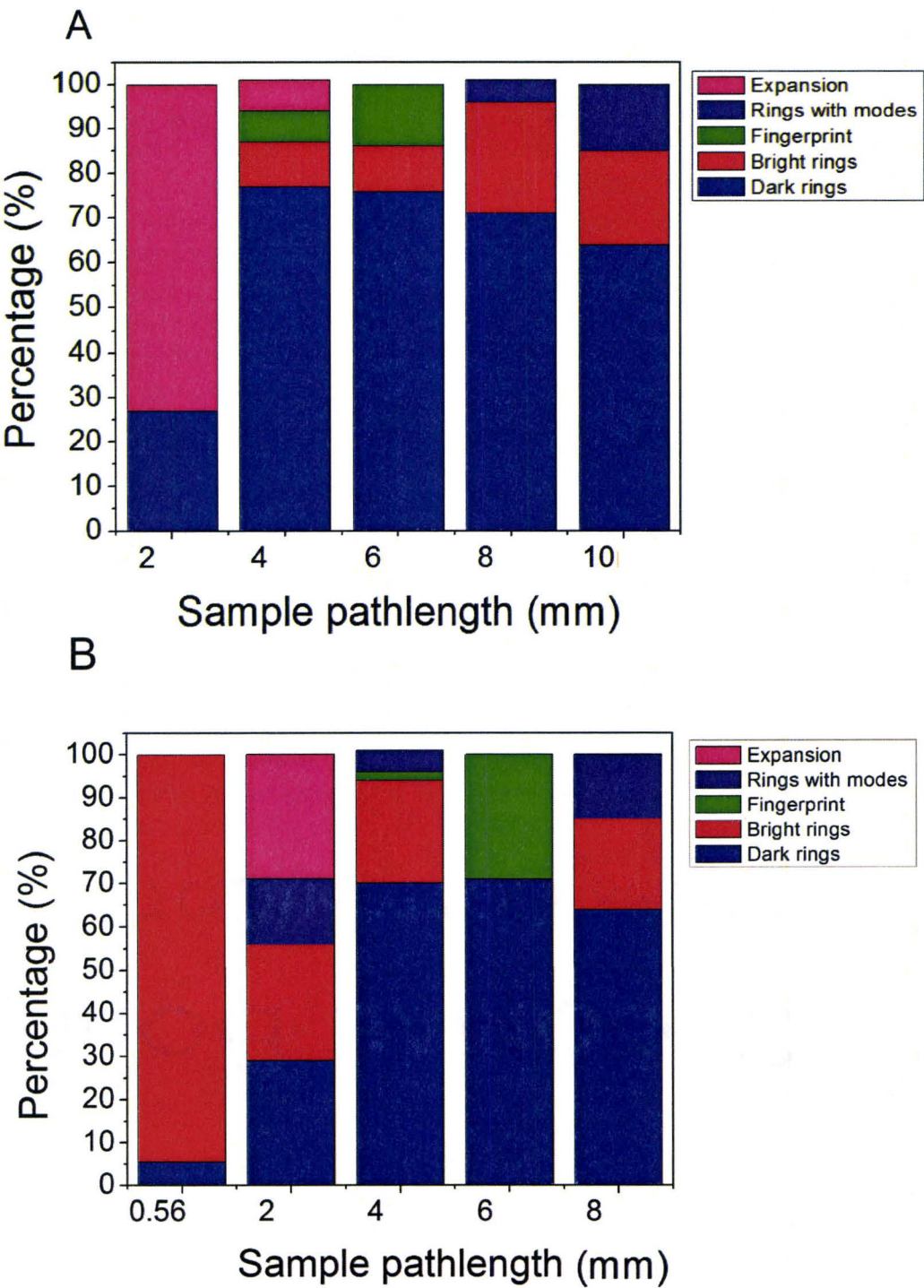


Figure 4.18: Statistics of diffraction ring patterns and expansion of the beam observed for various sample pathlengths at an input intensity of 1.6 W/cm^2 for rings A) imaged at the exit face of the medium and B) imaged further away from the exit face at various distances.

4.19a), the maximum peak intensity of the profiles presents oscillations over time (Figure 4.21).

At early times, these oscillations are accompanied with drastic changes in intensity profiles, for example the first oscillation occurs between 13 s and 20 s. At the peaks of this oscillation we see a bright centre surrounded by a ring (Figures 4.19b and d), during the valley of the oscillation a high order mode (Figure 4.19c) is observed corresponding to LP_{11} . For the second oscillation, between 20 s and 25 s, the last peak presents a bright centre now surrounded with at least three rings (Figure 4.19g) and during the valley, the high-order modes corresponding to LP_{11} and LP_{02} (24 s) superpose with three rings (Figure 4.19f). From 29 s to 70 s, fast oscillations in intensity are observed (Figure 4.21) accompanied with observation of high-order modes superposed with rings (Figures 4.19 i, j, k, m, n, o). For comparison, intensity profiles of high-order modes resulting from simulations of propagation of light at 532 nm in an optical fiber with $\Delta n = 0.08$ and 10 μm beam diameter in Beamprop are shown in Figure 4.20.

From 75 s onwards, oscillations in intensity are slower (Figure 4.21) and the intensity profile does not change substantially. A bright centre surrounded by multiple rings is observed. At peaks most of the intensity concentrates on the central spot and during valleys the intensity along rings increases. From the beginning of the experiment, the central bright beam self-focuses at the same time as rings emerge, which is indicated by an increase in intensity at every oscillation as shown in the temporal plot of relative intensity in Figure 4.21a and a decrease in beam diameter of the central bright spot. The total increase in intensity was approximately 4.5-fold and the bright spot diameter decreased from 387 μm to 145 μm from 13 s to 162 s. Rings with high-order modes were observed for all OPLs except 0.82 mm between 5 to 15% of the times (Figure 4.18).

The dynamics of this type of rings resembles the self-focusing dynamics at low powers. In that case, oscillations occurred and a low order mode and high-order modes were observed at the peak and valley of the oscillations, respectively (refer to Chapter 3). Both self-phase modulation and self-trapping of the beam occur simultaneously in this type of rings.

The evolution of intensity profiles (Figure 4.19) is consistent with that in beam propagation simulations performed through BeamPropTM (Chapter 6). Here, we observed oscillations of the beam with the formation of a bright centre surrounded by rings during the peak of the oscillation and high-order modes superposed with rings during the valleys (Figures 6.16 and 6.18). Self-focusing was indicated by the increase in intensity and narrowing of the beam at some of the peaks during oscillations. The refractive index induced in these simulations presents changes along all the propagating direction z since early times (Figure 6.19). This contrasts with simulations for dark rings where refractive index changes at early times only occurred at the entrance face (Figure 6.25).

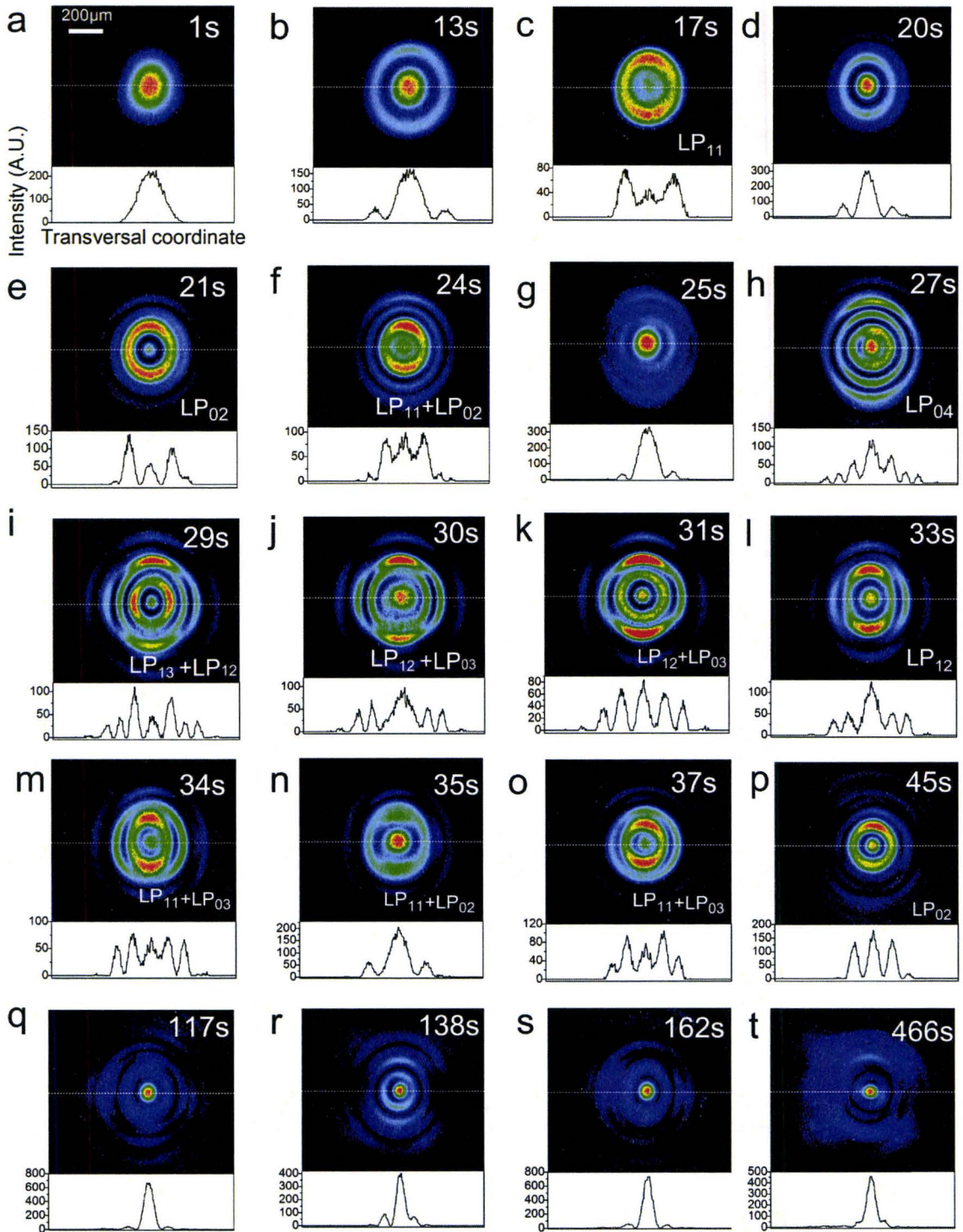


Figure 4.19: Temporal evolution of diffraction rings with high-order modes for an intensity of 1.6 W/cm^2 and an optical path length of 2.94 mm imaged at 10 mm from the exit face. 2-D and 1-D spatial intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value

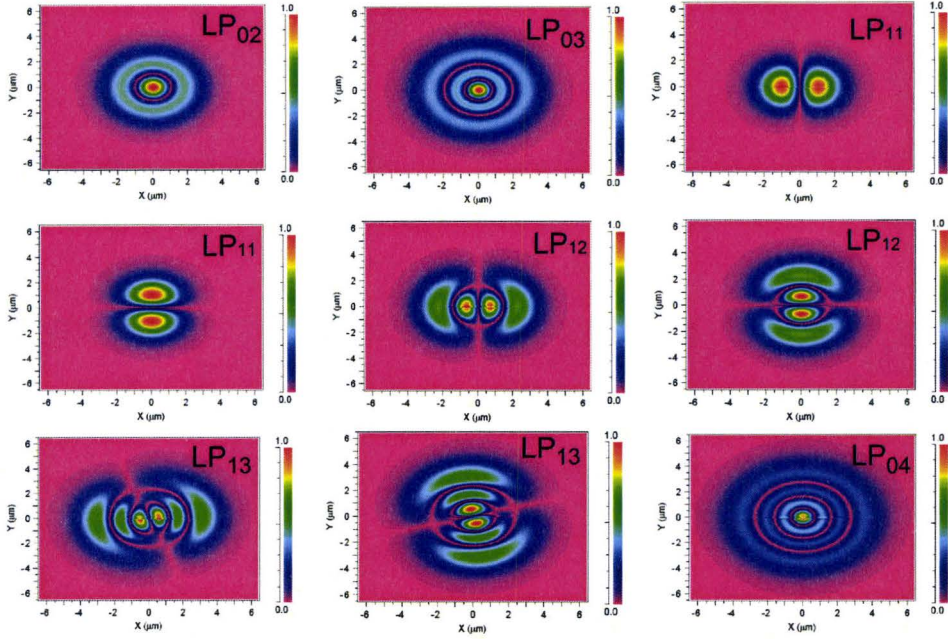


Figure 4.20: Beam propagation simulations of linearly polarized modes in an optical fiber performed with BeamPROP software. Modes were calculated for an optical fiber with diameter=10 μm and a refractive index contrast of 0.08.

A possible explanation for the observation of rings with high-order modes is that the degree of pre-polymerization of the sample is small compared to samples where dark rings were observed. For the latter, the maximum possible change (Δn_s) in refractive index is therefore smaller, saturation is achieved at earlier times and leads to a top-hat refractive index profile. Self-focusing is not as efficient and only diffraction rings are observed initially with the beam self-focusing at later times. In contrast, a system with a smaller degree of pre-polymerization has a greater Δn_s and therefore saturates slowly. Based on the kinetics of free-radical polymerisation, the less the extent of polymerisation the greater the rate of polymerisation. The beam under these conditions, is able to induce a Gaussian index profile which focuses light more efficiently and therefore faster, resulting in the observation of self-focusing at earlier times superposed with diffraction rings. Self-trapping at the low intensity regime was accompanied by oscillations and high-order modes, therefore we also observed both as seen in Figures 4.19 and 4.21.

Fingerprint rings: They had the characteristic of rings emerging all at the same time, resembling a fingerprint. This contrasts with the dark rings where rings emerged sequentially over time. The temporal evolution of fingerprint rings, which shown in Figure 4.22, it occurred as follows: in the first second the Gaussian beam diffracted to a beam diameter of 120 μm at the exit face of the sample (Figure 4.22 a). This was followed by a flattening of the profile occurring at 6s (Figure 4.22 b) and the emergence of multiple rings all at the same time in the next 2 s (Figure 4.22 c).

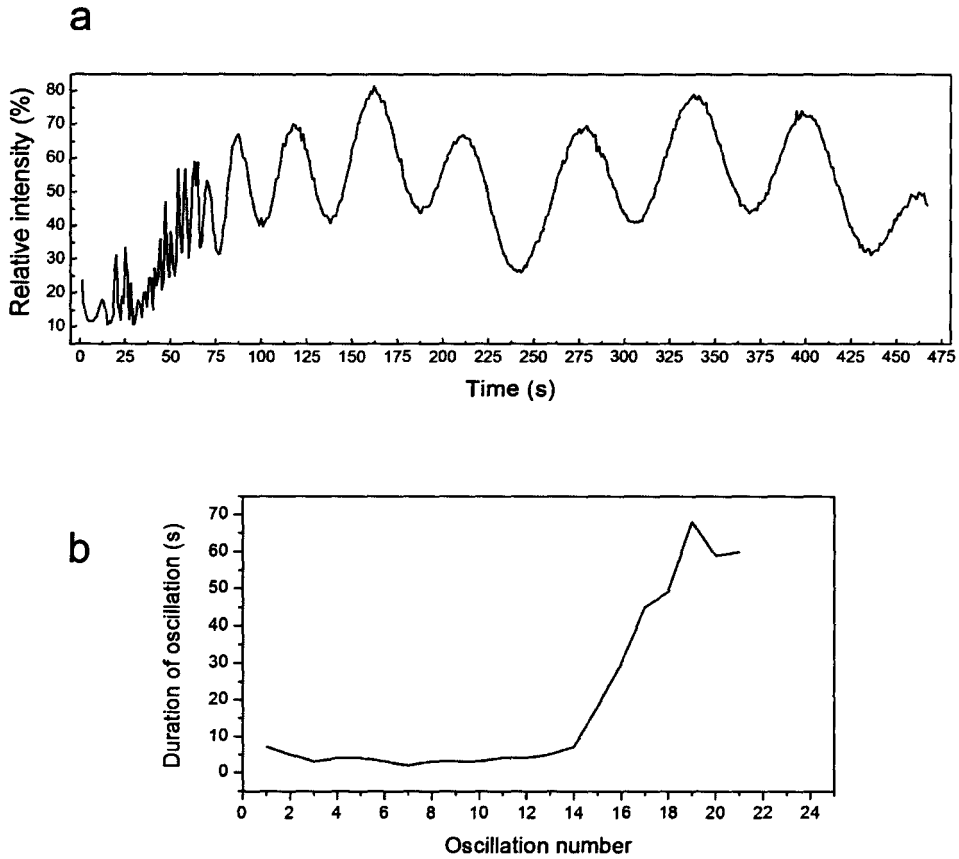


Figure 4.21: a) Relative maximum intensity (of the maximum CCD camera response) of the spatial profile as a function of time for diffraction rings with high-order modes at an intensity of 1.6 W/cm^2 and an optical path length of 2.94 mm imaged at 10 mm from the exit face and the corresponding b) duration of oscillations.

The rings increased in intensity with time (Figures 4.22 d-g) until the entire beam self-trapped into a narrow (25 μm) beam (Figure 4.22 i).

Having all the rings emerging at once and not changing in number over time, means that the maximum refractive index induced Δn_{max} and therefore the maximum phase shift $\Delta\psi_0$ affecting ring formation are reached immediately after exposure to the beam. This implies that the initial refractive index of the prepolymerized sample was greater compared to that where dark rings were observed. Therefore, the refractive index was closer to saturation, at which point polymerization reactions slowed down and the intensity profile did not exhibit significant changes. The maximum number of rings observed for fingerprint rings was 3 and 10 while for dark rings was 5 and 19 rings, for OPL= 5.88 mm and 8.82 mm, respectively. This supports our proposed mechanism since less rings would be induced for a smaller Δn_{max} .

Fingerprint rings were only observed for optical path lengths of 5.88 mm and 8.82 mm, from 2 % to 7 % in the former case to 14 % to 29 % of the times in the latter case.

Bright rings: Rings with a bright central spot have been predicted and observed only when the beam has a positive curvature at the entrance face of a self-focusing medium [85; 83]. Their observation in the organosiloxane with the entrance face of the sample at the beam waist could be the result of the sample being positioned outside of the Rayleigh range, with the beam having a small positive radius of curvature. The dynamics of bright rings observed in this case are the same as those observed for positive curvatures in the previous Section. That is, diffraction rings developed surrounding a bright centre and exchange of intensity occurred between the rings and the bright centre. In Figure 4.18, statistics of ring patterns for various pathlengths show that bright rings were only observed between 10% and 29% of the times for OPL > 2.94 mm. However, for the shortest OPL of 0.82 mm, bright rings were observed 94% of the times when imaged further away from the output of the sample (Figure 4.18b). Possibly due to the small thickness of the sample, there was a significant error (± 0.25 mm) in positioning.

4.8 Pathlength dependence

In Section 4.3, we monitored the evolution of dark rings at the output of the sample which had an optical path length (OPL) of 8.82 mm, the OPL being approximately ten times the Rayleigh range (z_R) of the focused beam. We assumed that initially most of the refractive index changes contributing towards self-phase modulation occurred at $OPL < z_R$ in the organosiloxane and propagation thereafter was linear. We partially confirmed this by repeating the experiment under similar conditions with a thin sample of OPL=0.82 mm ($\sim z_R$) obtaining a similar number of rings at the far-field. In this Section, we examine further the propagation of dark diffraction rings along the organosiloxane by performing experiments at an intensity of 1.6 W/cm² under

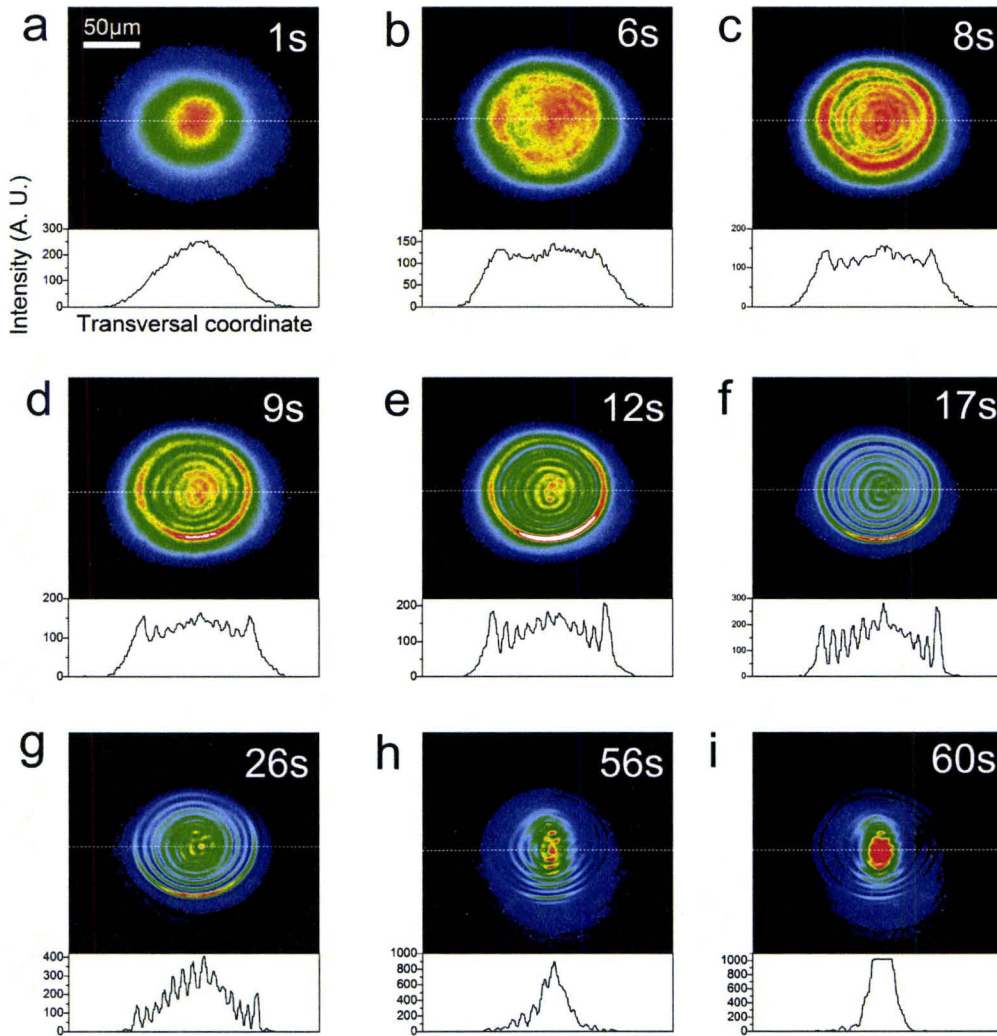


Figure 4.22: Temporal evolution of fingerprint diffraction rings for an intensity of 1.6 W/cm^2 at an optical pathlength of 5.88 mm imaged at the exit face. 2-D and 1-D intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value

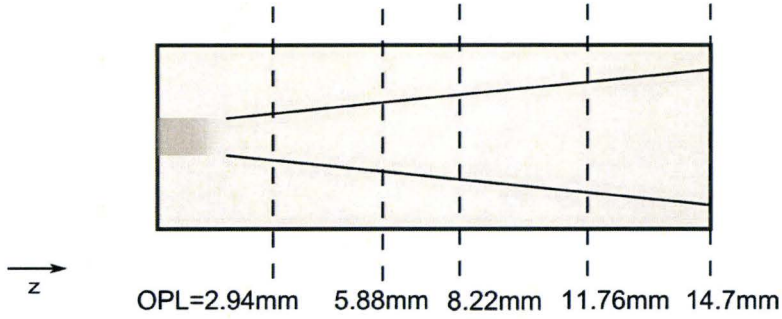


Figure 4.23: Scheme of pathlength dependence study. The propagation of diffraction rings is monitored at the output face of samples with different OPLs in order to investigate dynamics. The dark grey square at the entrance face of the sample represents the area of refractive index changes that mostly contribute to diffraction ring formation.

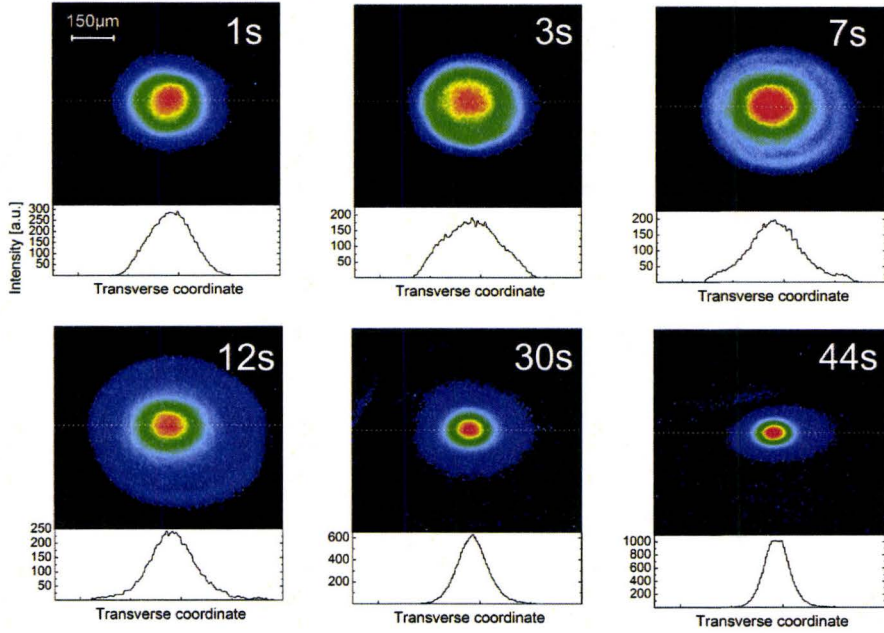
similar conditions as in Section 4.3 for various OPLs including 2.94 mm, 5.88 mm, 8.82mm, 11.76 mm and 14.7 mm (Figure 4.23). We monitored the intensity profiles at the output of the medium and further away in some cases to confirmed that the propagation of rings after z_R is linear and consistent with our proposed mechanism in Section 4.3.

Diffraction can be divided into near-field and far-field (Fraunhofer) diffraction [34]. The former occurs close enough to the entrance face that the curvature of the wavefront must be taken into account, as a result, the shape and size of the diffraction pattern changes with observation distance in the near-field. Far-field diffraction occurs far enough from the entrance face that wavefronts at the observation plane may be considered to be planar. In this case, the diffraction pattern changes monotonically in size with observation distance. Therefore, if changes that elicit self-phase modulation occur only at the entrance of the sample, the number of rings observed in the far-field should be the same for all OPLs.

By analysing the resulting spatial intensity profiles monitored at the output of the medium, we observe differences at the various OPLs. For short distances, including OPL=2.94 mm and 5.88 mm, we see a central bright beam present most of the time with a maximum ring formation of 1 and 5 rings, respectively (Figure 4.24 and Table 4.2). At longer OPLs we do not observe a central bright beam, for OPLs =8.82 mm and 11.76 mm (Figures 4.2 and 4.25a), multiple dark rings emerge as described in Section 4.3 with a maximum of 19 rings (Table 4.2). At an even longer OPL=14.7 mm, a maximum number of 5 rings appear. As the first ring forms at 4s (Figure 4.25b) it starts filamenting and in the next ten seconds multiple rings continue to emerge as filamentation occurs from the outermost ring to the inner ones at 15s (Figure 4.25b).

At all OPLs self-focusing occurs but at different timescales as shown in Table 4.2. At short OPLs=2.94 mm and 5.88 mm it occurs at around 45 s, for the longer OPLs=8.82 mm and 11.76 mm it takes place in the hundreds of seconds and then it

a) OPL=2.94 mm



b) OPL=5.88 mm

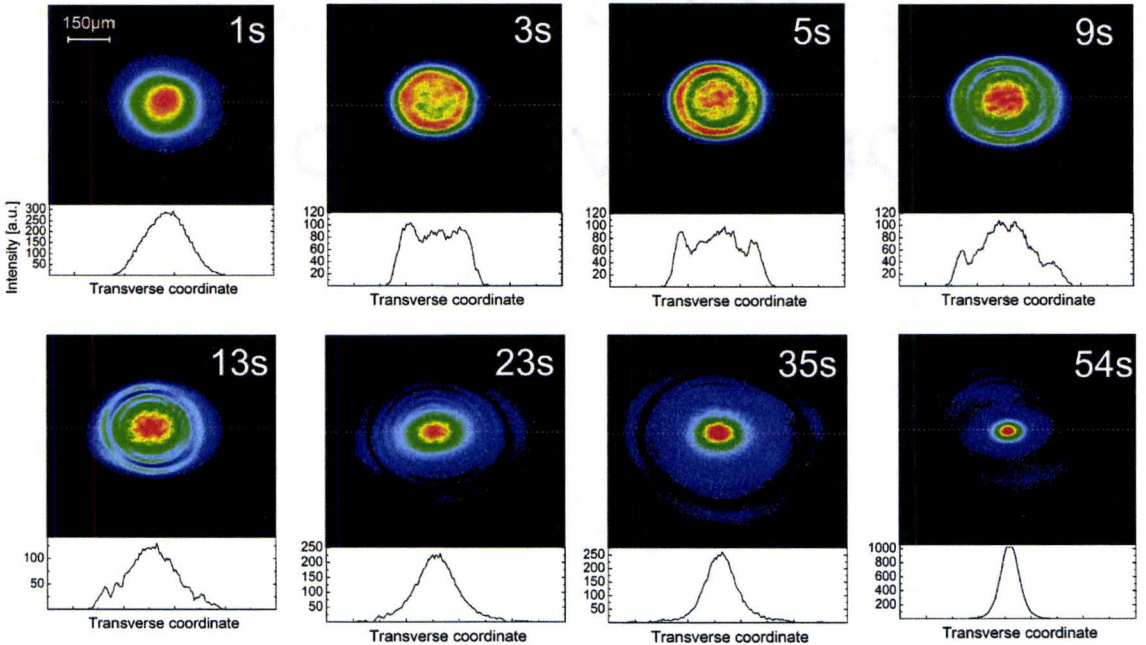
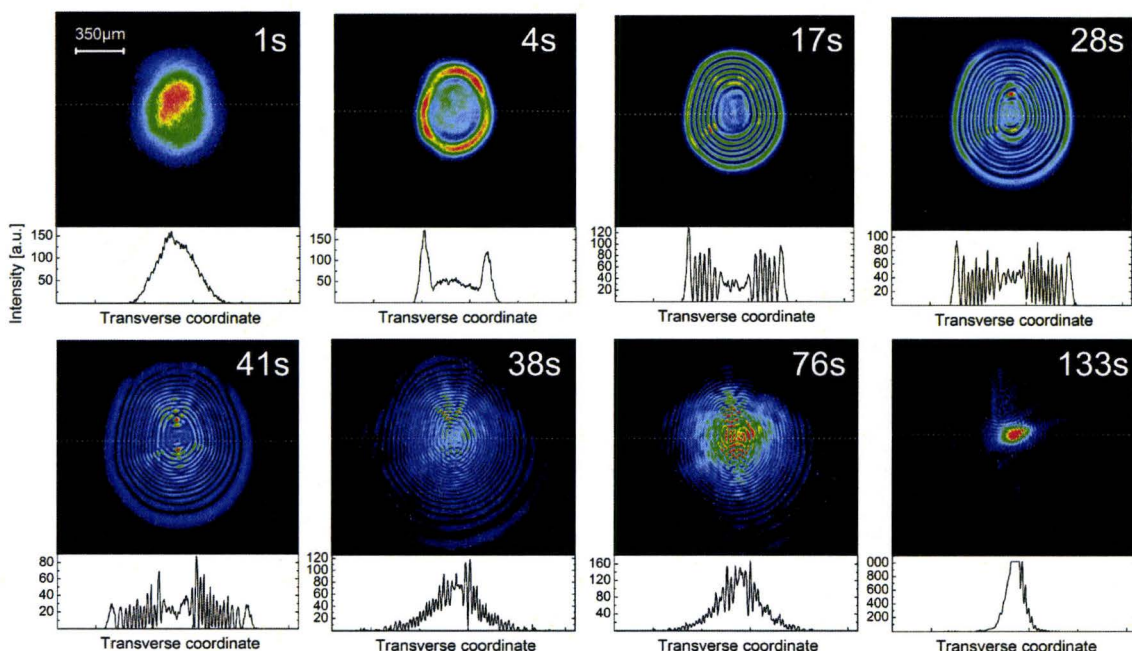


Figure 4.24: Temporal evolution of diffraction ring formation at an intensity of 1.6 W/cm² for a) OPL=2.94 mm and b) OPL=5.88 mm monitored at the output of the medium. 2-D and 1-D spatial intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value.

a) OPL=11.76 mm



b) OPL=14.7 mm

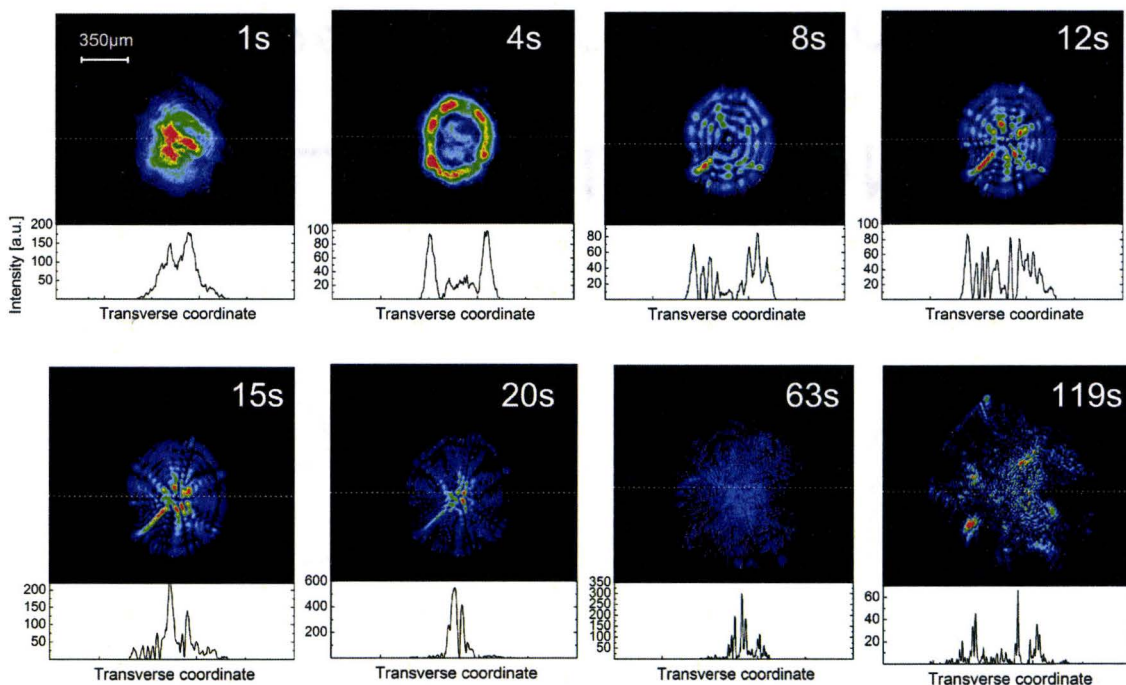


Figure 4.25: Temporal evolution of diffraction ring formation at an intensity of 1.6 W/cm^2 for a) $\text{OPL}=11.76 \text{ mm}$ and b) $\text{OPL}=14.7 \text{ mm}$ monitored at the output of the medium. 2-D and 1-D spatial intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value.

Table 4.2: Statistics of pathlength dependence parameters for dark rings

OPL (mm)	Max.# rings	Avg. diameter of first ring (μm)	Stdv.	Avg. time of first ring (s)	Stdv.	Avg. self-focusing time (s)	Stdv.
2.94	1	112	6	9	3	48	6
5.88	5	114	34	11	10	46	15
8.82	19	132	26	4	2	111	42
11.76	19	165	40	5	2	87	39
14.7	5	143	35	3	0	26	10

Table 4.3: Statistics of pathlength dependence parameters for dark rings far-field

OPL (mm)	Max.# rings	Avg. time of first ring Initial (s)	Stdv.	Avg. Self-focusing time (s)	Stdv.
2.94	6	6	2	68	24
5.88	8	6	5	79	42

decreases to around 26 s for the longest OPL=14.7 mm. At this pathlength, even if filamentation occurs, at 20 s the beam self-focuses indicated by the 3-fold increase in intensity, the beam narrows from $\approx 440 \mu\text{m}$ to $263 \mu\text{m}$ although filamentation is still present (Figure 4.25b). The resulting statistics for the average diameter of the first ring that emerges show that the diameter increases with increasing pathlength except for the longest OPL=14.7 mm. The average time of appearance of this ring is below 10 s for all OPLs and it seems to decrease with pathlength.

Assuming that the propagation is linear after z_R , we calculated the expected diameter of the first ring at different pathlengths by calculating the half-angle formed by the diffraction rings for an OPL=8.82. The half-angle resulted in 0.007 degrees and the diameter was $44 \mu\text{m}$, $88 \mu\text{m}$, $132 \mu\text{m}$, $176 \mu\text{m}$ and $220 \mu\text{m}$ corresponding to the pathlengths 2.94 mm, 5.88 mm, 8.82 mm, 11.76 mm and 14.7 mm, respectively. By comparing these values with the diameters measured in experiments for the various pathlengths shown in Tables 4.2 and 4.3, the diameter trend only agrees for OPL=8.82 mm and 11.76 mm. For the two shorter pathlengths the diameter values are much larger than the expected ones and for the longest pathlength the diameter value is smaller.

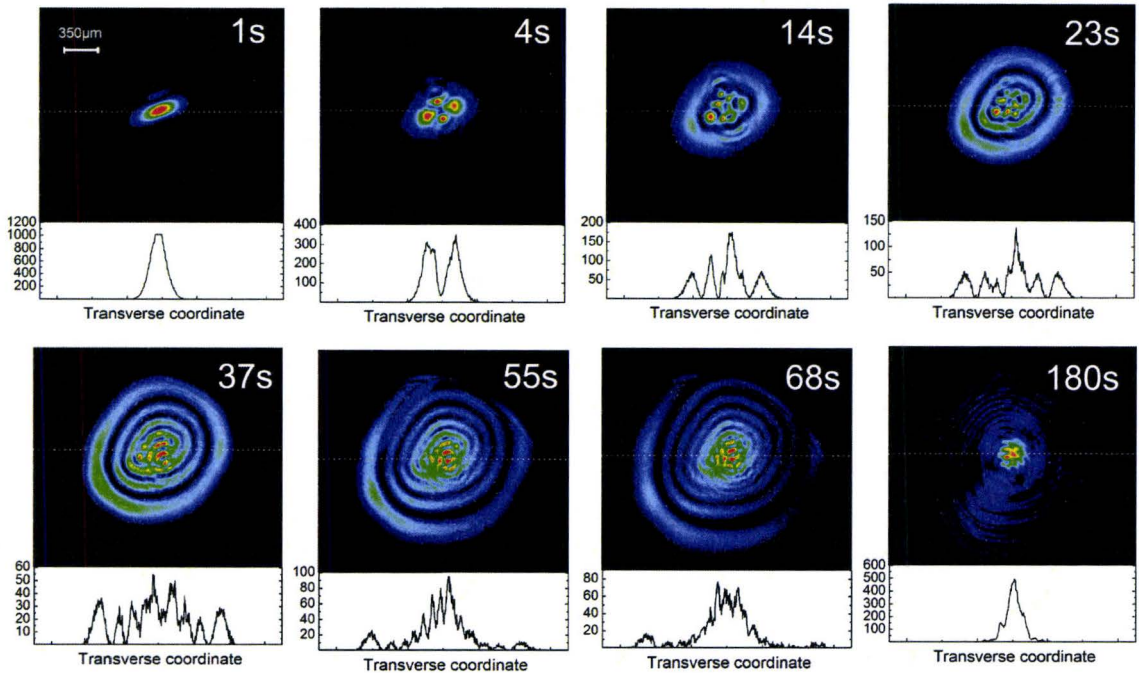
From the difference in diameter value and differences in spatial profiles at various OPLs, we deduce that at short pathlengths we were imaging at the near-field therefore number of rings and spatial profiles were different compared to longer pathlengths. Previous theoretical studies of ring formation in other nonlinear media predict varying patterns at the near field [88; 84]. To confirm this and obtain the maximum number

of rings at the far-field we performed experiments for short OPLs where we imaged further away from the exit face at $z > 10$ mm for OPL=2.94 mm and OPL=5.88 mm.

At the far-field, we obtained spatial profiles more similar to longer pathlengths with multiple ring formation and without the observation of the initial central bright beam (Figure 4.26). However, the maximum number of rings was 6 and 8 for an OPL=2.94 mm and OPL=5.88 mm respectively, which is below the 19 rings observed for the next two OPLs (Table 4.3). The apparent contradiction of these results with the fact that we observed in thin samples (OPL=0.82 mm) a similar number of rings as in samples with OPL=8.82 mm (Section 4.3) can be explained by the interplay of self-focusing and self-phase modulation. In thin samples, self-focusing does not take place since $\text{OPL} < z_R$ and only the effects of spatial self-phase modulation are observed, however in samples with $\text{OPL} \geq z_R$ both self-focusing and self-phase modulation occur. From the different self-focusing times for different OPLs (Tables 4.2 and 4.3), self-focusing dynamics vary, for short OPLs (2.94 mm and 5.88 mm) this process occurs earlier which probably is related with the observation of less number of rings compared to longer OPLs (8.82 mm and 11.76 mm) where self-focusing occurs later and more rings are observed. For the longest OPL=14.7 mm, the dynamics are more complex as multiple phenomena occur simultaneously, (i) self-phase modulation giving rise to ring formation, (ii) modulation instability probably triggered by the long OPL, which seeds noise in the system resulting in filamentation and finally (iii) self-focusing of the beam. Previous theoretical studies have shown the filamentation of rings as a function of propagation distance [113], rendering them unstable at large pathlengths to noise in the system ultimately breaking into individual filaments.

From the above results, changes in refractive index contributing to self-phase modulation very likely occur at least up to 5.88 mm. The propagation of diffraction rings then seems to follow linear propagation up to OPL=11.76 mm. At the near field, the shape and size of the diffraction pattern changed with observation distance and at the far-field the diffraction rings increased monotonically in size (according to linear diffraction). However, at very long pathlengths filamentation of the rings disrupts the linear propagation. The disagreement in the total number of rings obtained at the far-field for different OPLs is related with the different self-focusing dynamics. At the shorter OPL=2.94 mm and 5.88 mm and the longest OPL=14.7 mm, self-focusing occurs earlier compared to OPL=8.82 mm and 11.76 mm. Therefore, less number of rings are observed as self-focusing disrupts the observation of diffraction rings. That the average time of appearance of the first ring is below 10 s for all OPLs, means that refractive index changes occur in the same timescale for all OPLs. This further supports the notion that refractive index changes contributing to self-phase modulation originates mainly from the entrance face. Simulations for dark rings from Section 6.4.2 agree with our conclusions that refractive index changes for early times occur mainly at the entrance face.

a) OPL=2.94 mm far-field



b) OPL=5.88 mm far-field

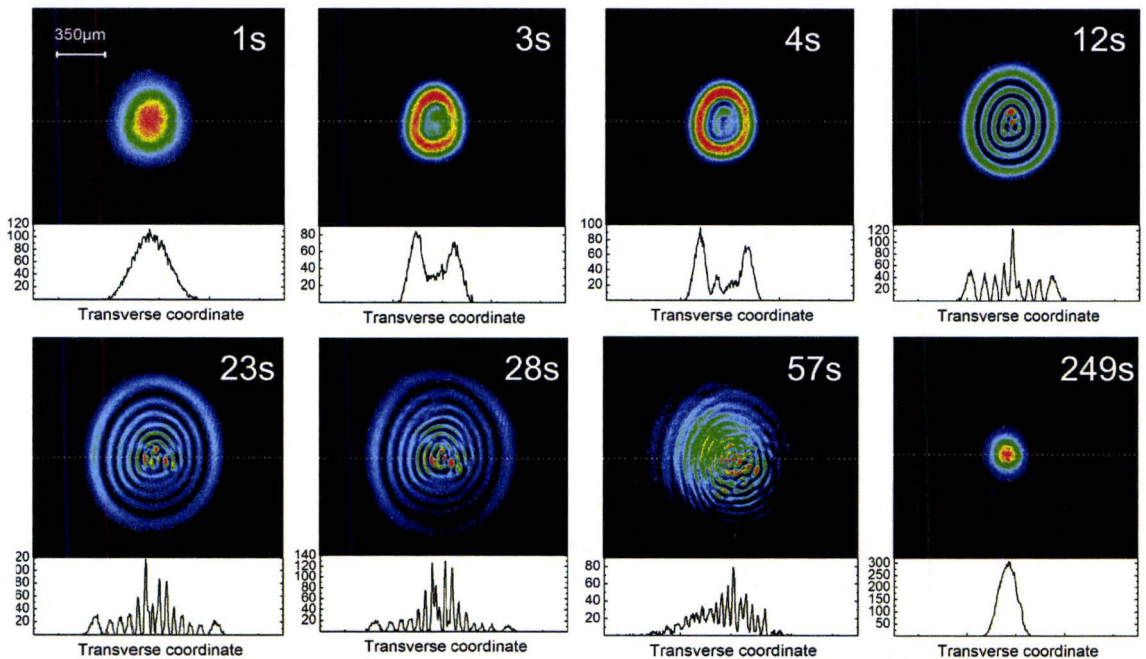


Figure 4.26: Temporal evolution of diffraction ring formation at an intensity of 1.6 W/cm^2 for a) OPL=2.94 mm and b) OPL=5.88 mm monitored at the far-field. 2-D and 1-D spatial intensity profiles of the beam are shown. For clarity, each 2-D profile has been normalized to the maximum intensity value.

Table 4.4: Statistics of dependence on pre-polymerization

Pre-polymerization time (min)	Max.# rings	Avg. Self-focusing time (s)	Stdv.
5	3	didn't occur	
10	10	115	39
15	5	48	1
20	2	46	6

4.9 Dependence on pre-polymerisation of organosiloxane

Prior to experiments on diffraction rings induced by a laser beam, samples were uniformly polymerized with white light as detailed in Section 2.2. The duration of uniform irradiation determines the degree of polymerisation in the organosiloxane and hence the initial background refractive index and thus Δn_s [39]. Because the emergence of diffraction rings and their number relies on the maximum phase shift $\Delta \Psi_0$ induced by the refractive index profile, the maximum possible refractive index change in the sample (Δn_s) is of critical importance to be able to induce more or less rings.

In order to investigate the impact of degree of polymerization in the emergence of diffraction rings, we pre-polymerized samples with OPL=8.82 mm for 5, 10, 15 and 20 minutes prior to laser exposure at 1.6 W/cm². The maximum number of rings induced and the self-focusing time varied for the different exposure times as shown in Table 4.4. An optimum polymerization time of 10 minutes was found to maximize the number of rings and delay self-focusing time to 115 s. For the low pre-polymerization time of 5 minutes, self-focusing did not occur and for longer exposure times of 15 and 20 minutes, self-focusing took place at approximately half of the time of the 10 minute exposure samples.

These results indicate that at low pre-polymerization times, the organosiloxane matrix does not have sufficient density to support self-trapping of the beam. That means that an induced waveguide cannot form as the matrix is not sufficiently pre-polymerized. However rings are observed and this could be the result of refractive index changes occurring only at the entrance face. The small number of rings possibly results from the slow changes in refractive index. At high exposure times, few methacrylate units are available to polymerize in the organosiloxane and therefore the remaining refractive index change is not enough to induce a greater number of rings.

4.10 Permanent structures after diffraction rings

The refractive index changes responsible for self-phase modulation and diffraction ring formation in the organosiloxane were permanent and provided information on the propagation path of the diffraction rings through the sample.

Figure 4.27 A, corresponds to the optical micrograph of a permanent conical structure resulting after the observation of dark ring in sample with OPL=8.82 mm, which was acquired after the emergence of six diffraction rings (at 14 s) in the organosiloxane (shown in the left of Figure 4.27 A). The micrograph reveals index changes due to polymerization that were permanently inscribed in the medium during self-phase modulation. The resulting conical microstructure corresponds to the trajectory of the interference cones from the entrance face to the exit face of the medium. Accordingly, the apex of the cone is located near the entrance face while its lateral surface corresponds to the most intense outermost ring.

By contrast, the micrograph acquired after only self-focusing and self-trapping of the beam (at the lower intensity of 0.016 W/cm^2 where self-phase modulation is not possible) contains a narrow cylindrical waveguide with uniform diameter maintained along the propagation axis (Figure 4.27 C).

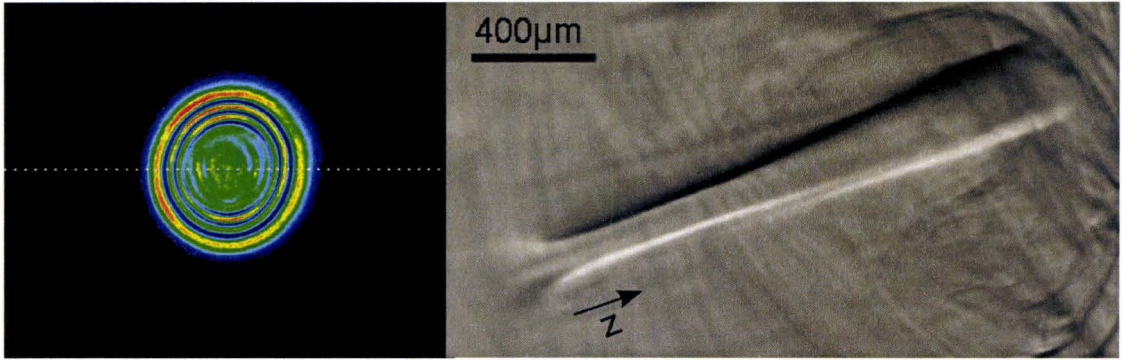
The emergence of dark rings was followed by self-focusing of the beam and in some cases we were able to observe an inner waveguide formed surrounded by the conical microstructure as shown in Figure 4.28. In this case, the surrounding conical microstructure corresponds to the emergence of 14 diffraction rings at 30 s and the inner waveguide to the self-focusing of the beam at 150 s.

Figure 4.27B, shows the micrograph of a permanent structure inscribed by bright diffraction rings in a sample with OPL=8.82 mm. The microstructure has a conical shape similar to the one observed after the emergence of dark rings, however the width of the cone at the entrance and at the exit face is at least 3 times smaller. This is explained by the fact that most of the intensity was localised in the bright central beam and the first two diffraction rings during the emergence of rings (see Section 4.7).

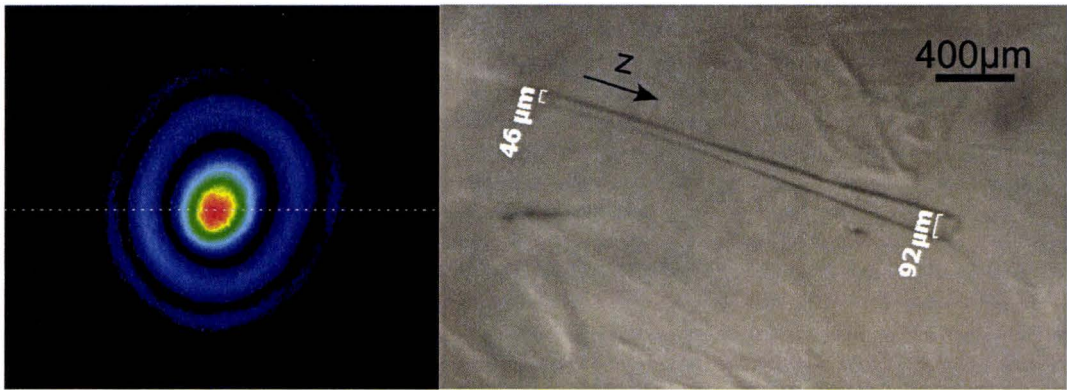
4.11 Conclusions

In this Chapter, the dynamics of diffraction rings induced through laser initiated free-radical polymerization were demonstrated for the first time. Propagation of diffraction rings over long distances $\gg$ Rayleigh range (z_R) was shown, which differs from most previous studies where thin samples ($< z_R$) were employed to prevent simultaneous self-focusing of the beam. Monitoring of the temporal evolution of diffraction rings *in situ* was possible as the organosiloxane presents a slow photoresponse. Through this study, parameters such as number of rings and ring diameter were extracted over time, which allowed to link polymerisation kinetics to the evolution of

A. Dark rings



B. Bright rings



C. Only self-trapping



Figure 4.27: Optical micrograph of permanent conical structures formed after A) dark diffraction rings and B) bright diffraction rings in the right hand side and their corresponding intensity profiles in the left hand side for a sample ($\text{OPL} = 8.82 \text{ mm}$) with 1.6 W/cm^2 of input beam intensity. For comparison, the permanent waveguide structure induced by a self-trapped beam is shown (C). The propagation direction of the beam (z) is indicated. (In order to best visualize the structures, micrographs in (A) and (B) were acquired with the sample placed at an angle. The scale bar is therefore approximate).

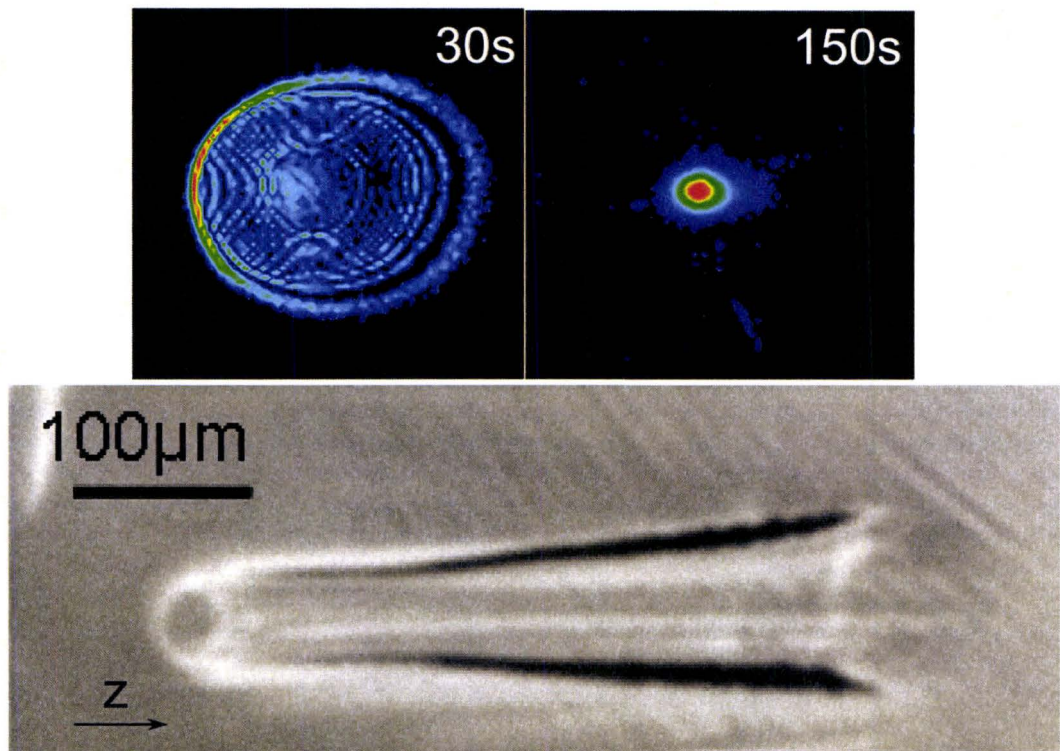


Figure 4.28: Optical micrograph of a conical permanent structure with an inner waveguide formed after the observation of diffraction rings followed by self-trapping. The corresponding intensity profiles are shown in the top for a sample with OPL = 8.82 mm at 1.6 W/cm². The propagation direction of the beam (z) is indicated.

diffraction rings. A mechanism where significant refractive index changes occur at the entrance face of the sample and linear propagation of diffraction rings occurs through the sample was proposed. Studies of the pathlength dependence of self-phase modulation revealed that at short pathlengths the propagation corresponded to near-field diffraction, at longer pathlengths to far-field diffraction and at the longest pathlength, propagation was disrupted with filamentation of the diffraction rings.

Studies of the dependence of self-phase modulation on beam curvature were performed in long pathlengths. Our results showed that rings with a dark centre are induced at $R < 0$ and rings with a bright centre are most of the times induced at $R > 0$. This overall result agrees with rings induced in thin samples. However in our studies we were able to observe dynamics of diffraction rings in a long pathlength medium. Dark rings induced at $R < 0$ showed filamentation with time and an increase of the dark centre diameter. The former showed that the medium became susceptible to noise when self-trapping occurred at longer times and no significant changes took place in the induced diffraction rings after some time. The latter was found to be the result of an effective increase in beam diameter for the contribution of refractive index changes to the total phase shift. Bright rings showed an exchange of intensity between the central bright beam and the first two rings. Unlike other media, the organosiloxane underwent permanent refractive index changes. Structures induced by dark and bright rings were characterised after their formation, which allowed for the visualization of nonlinear light propagation in the medium.

Through careful analysis of statistics of experiments, different diffraction rings were observed at $R = \infty$ including dark rings, rings with high-order modes, fingerprint rings and bright rings. This variety of ring patterns has not been observed before. The observation of rings with high-order modes and fingerprint rings were attributed to slightly lower and slightly higher pre-polymerization of the samples, respectively.

Pathlength dependence studies were performed and the observed dynamics indicated that light propagation is more complex than expected. Self-trapping times varied with propagation length and this affected the observed number of diffraction rings. A linear propagation with a contribution at the entrance of the sample was found for the OPL=8.82 mm and 11.76 mm. At longer pathlengths, diffraction ring emergence was accompanied with filamentation of the rings.

Chapter 5

Formation and spontaneous filamentation of a single optical ring originating from an input Gaussian beam

5.1 Introduction

Optical beams in nonlinear systems spontaneously break up in multiple filaments at high powers [96; 95; 102]. This effect originating from transverse modulation instability in the beam has been observed in whole beams and in ring-shaped beams [128; 102]. Input super-Gaussian (SG) beams propagating in Kerr media, have been observed to collapse into a ring profile (rather than the Townes profile) at high powers and further filament due to azimuthal perturbations ultimately forming a ring of filaments [99]. In this study, we show that an input Gaussian beam in an organosiloxane can evolve into a ring-shape beam when propagating under nonlinear conditions and later suffer filamentation. This is fundamentally different from what occurs in Kerr media where the instantaneous and localized nonlinearity requires an input SG beam, generated with spatial modulation optics of ultra-short laser pulses (on the order of femtoseconds) with intensities on the order of 10^{10} W/cm² to collapse into a ring profile at the exit of the cell. In contrast, the organosiloxane possesses a saturable nonlinearity that allows for an input Gaussian beam to induce a flattened Gaussian refractive index profile in the medium that evolves into a single ring at intensities as small as 27 W/cm² using a continuous wave laser.

Filamentation of the single ring in the organosiloxane is similar to filamentation of rings resulting from super-Gaussian beams in Kerr media and of rings with uniform phase in that noise seeds the breakage of the ring [99; 112]. Other ring-shape beams such as optical vortices which are ring-shaped profiles with helical phase in Kerr media, filament at high power due to asymmetric phase [100]. The unique feature of filamentation in the organosiloxane is that we are able to monitor the filamentation process over time due to the slow changes in refractive index. This is not possible in Kerr media as changes occur instantaneously and the only way to monitor changes is to perform experiments at different sample pathlengths [99].

In the following Chapter, first we show experimental results of the dynamics of the Gaussian beam at an input intensity large enough to be able to induce such

a refractive index profile where single-ring formation and further filamentation are observed. Then we prove that a flattened refractive index profile can be induced in our saturable system based on free-radical photopolymerization. We then provide a comparison between our experimental results and nonlinear simulations performed with BeamPROPTM for single ring formation. We show the filamentation dependence with input intensity experimentally and finally we show the structures formed in the photopolymer as a result of the permanent refractive index changes suffered during nonlinear light propagation.

5.2 Transition of a Gaussian beam from diffraction rings to a single-ring profile and its filamentation

The intensity dependence study performed in Chapter 3 showed that in the organosiloxane, self-trapping along with the observation of oscillations and high-order modes occurred at low intensities (3.2×10^{-5} W/cm² to 0.016 W/cm²). As the input intensity was increased in the system, the initial refractive index induced was greater and the corresponding index profile allowed for diffraction rings to be observed. At even greater intensities, the lack of sufficient index contrast in the induced profile only permitted filamentation of the whole beam to occur. In this section we show that at input intensities from 27 W/cm² to 111 W/cm², midway between the occurrence of diffraction rings and filamentation, a different phenomenon emerges. We first observe the effects of self-phase modulation with the emergence of diffraction rings followed by the formation of a single ring. Subsequently, due to modulation instability, filamentation of the single ring occurs. These phenomena are possible due to the saturability of our medium, the slow changes in refractive index and the fact of being at an intensity range where the index profile transforms from Gaussian to flattened Gaussian within seconds.

Preparation of the organosiloxane photopolymer employed in this study has been detailed elsewhere (Sections 2.2 and 2.3). Briefly, the organosiloxane consisted of a sol of oligomeric siloxanes with photopolymerizable methacrylate substituents. The sol was sensitized to visible wavelengths through addition of 0.05 wt% of the titanocene free-radical photoinitiator ($\lambda_{max} = 393$ nm, 460 nm). The sol was contained in a cylindrical cell ($d = 12$ mm, pathlength $z = 6.0$ mm) with optically flat and transparent windows. The sample was first partially photopolymerised and transformed into a gel through uniform illumination with white light emitted by a quartz-tungsten halogen lamp. For experiments, we focused a linearly polarized, continuous wave, 532 nm laser beam to a diameter ($1/e^2$) of 20 μ m and intensity of 80 W/cm² onto the entrance face ($z = 0.0$ mm) of the gel. The spatial intensity profile of the beam at the exit face ($z = 6.0$ mm) of the sample was imaged through a pair of planoconvex lenses onto a CCD camera and monitored over time.

Typical experimental results are presented in Figure 5.1. The beam initially diverged from a width of $20\text{ }\mu\text{m}$ at $z = 0.0\text{ mm}$ to $319\text{ }\mu\text{m}$ at $z = 6.0\text{ mm}$ (Figure 5.1 a-b).

As the beam initiated polymerization within the next few seconds, concentric rings emerged around the central beam and rapidly increased in number (Figure 5.1 c-e). Up to 5 such diffraction rings, the outermost of which was the most intense, could be resolved. With time, the diffraction rings decreased in intensity as the central beam increased slightly in intensity. The beam then developed a dark central spot that gradually widened, transforming the beam at 37 s into a single ring with a thickness of $190\text{ }\mu\text{m}$ and diameter of $420\text{ }\mu\text{m}$. At 61 s, the dark spot had widened sufficiently to decrease the thickness of the single ring to $27\text{ }\mu\text{m}$; the narrowing of the ring was accompanied by a 2-fold increase in relative intensity. Although the diffraction rings were faintly visible during this time, they underwent no further significant changes and eventually disappeared. The single ring, which maintained its diameter of $420\text{ }\mu\text{m}$ from this point, was unstable under the nonlinear conditions elicited by polymerization. Random variations in its intensity distribution became amplified; regions of even slightly greater intensity grew rapidly by depleting intensity from less intense regions. This modulation instability of the ring led to its spontaneous rupture into multiple filaments of light. Up to 17 filaments were observed at 100 s but these suffered rapid and random fluctuations in number and intensity (Figure 5.1i) until they stabilized at 253 s into a set of 7 azimuthally positioned filaments. Each filament exhibited strong self-trapping as indicated by a 20-fold increase in intensity (relative to the single ring at 61 s). The circular array of self-trapped filaments remained stable for as long as it was monitored (509 s) (Figure 5.1 k-l). The sequential transition of the Gaussian beam from diffraction rings into a single-ring structure, which stabilized upon filamentation was highly reproducible and observed in 5 of 5 experiments performed under identical conditions.

5.3 Evolution of induced refractive index profile: from Gaussian to flattened Gaussian

The behaviour of the beam in Figure 5.1 correlates directly to the spatial and temporal evolution of refractive index changes (Δn) that it induces in the organosiloxane. Index changes in the organosiloxane are large ($\Delta n = 0.006$), saturable and because they originate from a polymerisation reaction, they are slow, occurring over seconds to minutes [39]. Consequently, the spatial refractive index profile induced by the beam changes significantly over this timescale and in turn governs the nonlinear dynamics of the beam in the medium. The plot in Figure 5.2b traces the temporal evolution of the refractive index profile induced by the Gaussian beam at the entrance face (z

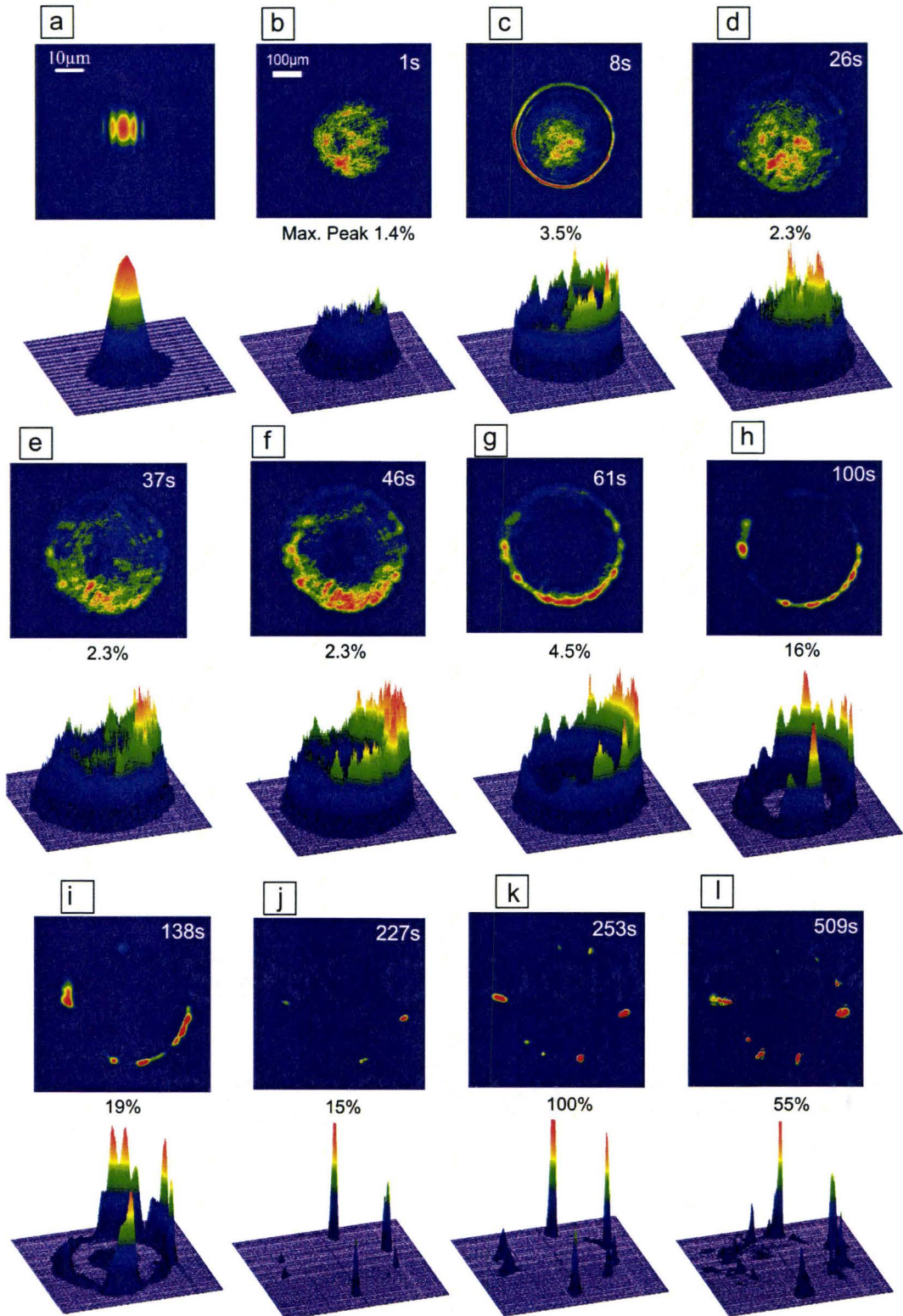


Figure 5.1: Temporal evolution of 2D and 3D intensity profiles of a laser beam with an input intensity of $80\text{W}/\text{cm}^2$ in a photopolymer. In (a) the input Gaussian beam is shown. Three main types of nonlinear propagation are observed: diffraction rings, single-ring formation and filamentation of the single-ring. 2D intensity profiles were scaled to peak for clarity.

= 0 mm) of the organosiloxane (Figure 5.2a). Profiles were calculated through the expression for index changes originating from free-radical polymerization [39]:

$$\Delta n(x, y, z, t) = \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(t)|^2 dt \right] \right\} \quad (5.1)$$

where Δn_s is the maximum index change (at saturation), U_0 , the critical intensity required to initiate polymerisation, τ , the monomer radical lifetime (assumed to be negligible) and $|E(t)|^2$, the square of the electric field amplitude of the incident optical field. $|E(t)|^2$ was replaced with the spatial intensity profile of a Gaussian beam,

$$I_r = I_{max} \exp \left(-\frac{2r^2}{\omega^2} \right) \quad (5.2)$$

where I_{max} is the intensity maximum, r , the radial coordinate and ω , the beam radius. Figure 5.2b shows that at a given intensity, the beam initially induces an index profile that corresponds to its own Gaussian shape (step 1).

Because the rate of polymerization is intensity-dependent, the maximum change in refractive index occurs at the most intense axial region with a cylindrically symmetric decay from this point. Polymerization and corresponding index changes also first reach saturation in the axial region (step 10). With time, as the less intense regions surrounding the axis also saturate, the refractive index assumes a flattened Gaussian profile (step 20). The evolution of the index profile from Gaussian to flattened Gaussian strongly modulates the nonlinear propagation of the beam. At early times, the Gaussian refractive index profile leads to self-phase modulation of the beam [6], which gives rise to the diffraction rings observed in Figure 5.1 c-e. Self-phase modulation occurs when the Gaussian index profile imposes a transverse phase shift on the beam according to [6]:

$$\Delta\psi(r) = \frac{2\pi}{\lambda} \int_{z_0}^{z_0+d} \Delta n(r, z) dz \quad (5.3)$$

where $\Delta n(r, z)$ is the refractive index change induced at a specific point in space, λ , is the free-space wavelength, z_0 , the entrance face along the propagation axis, and L , the propagation distance along which the beam acquires a transverse phase shift. According to Equation (5.3), the profile of the phase shift $\Delta\psi(r)$ will correspond to the Gaussian profile of the refractive index change ($\Delta n(r, z)$). Radiation from any two points along $\Delta\psi(r)$ with the same wavevector $k_\perp = (d\Delta\psi(r)/dr)$ will undergo constructive (destructive) interfere when $\Delta\psi(r_1) - \Delta\psi(r_2) = m\pi$, where m is an even (odd) integer. Multiple rings form when the maximum phase shift $\Delta\psi_0 > 2\pi$. The cylindrically symmetric profile of $\Delta\psi(r)$ produces a nested set of interference cones that propagate through the medium and are observed as concentric diffraction rings at $z = 6.00$ mm (Figure 5.1c). Consistent with the theory of self-phase modulation, the outermost ring, which originates from radiation with the greatest $k_\perp$ about the inflection point, $d\Delta\psi(r)/dr$ was the most intense (Figure 5.1c).

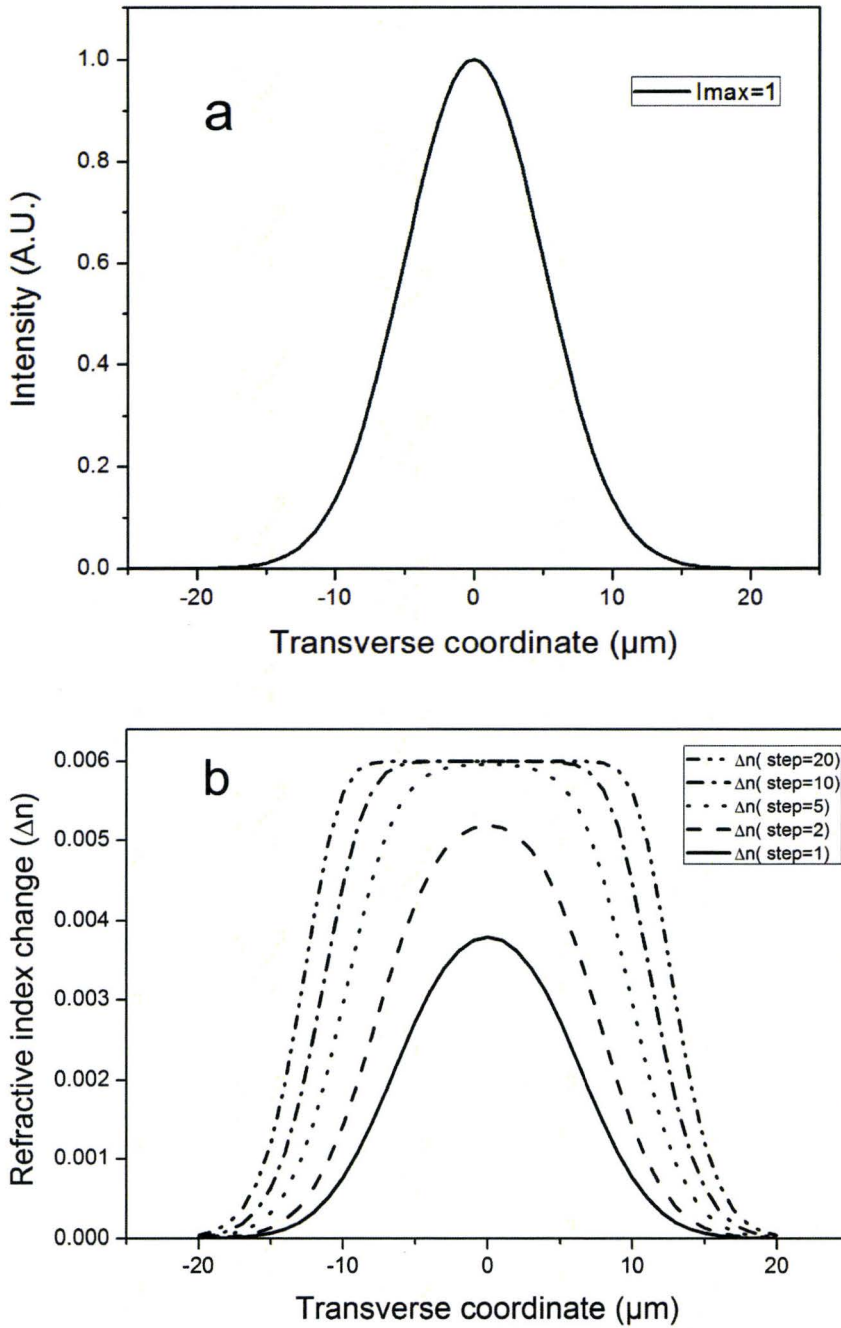


Figure 5.2: Plots of a) a Gaussian profile with $I_{max}=1$ and their corresponding b) refractive index profiles induced in the photopolymer where flattening of the Gaussian profiles is achieved at later steps. Each step represents the integrated intensity over 1, 2, 5, 10 and 20 unit times.

The transition of the beam from diffraction rings into a distinct single ring observed in Figure 5.1 d-g originates from the flattening of the Gaussian refractive index profile (Figure 5.2b). The beam encountering the flattened Gaussian index profile was focused into a ring that continued to propagate along z . We confirmed this through beam propagation simulations in which a Gaussian beam was launched into the photopolymer with a flattened Gaussian refractive index profile at the entrance face ($z = 0.0$ mm) [Section 5.4]. The beam propagated under nonlinear conditions and continued to induce refractive index changes along its propagation path according to Equation (5.1). Results presented in Figure 5.4 showed that under these conditions, the beam developed a dark central hole midway along its propagation path and was transformed into a ring. 1-D cross sections of the beam profile acquired at $z = 6.0$ mm showed excellent agreement with experiment (Figure 5.5).

Parallels exist between our findings and studies of a flat-top beam with a super-Gaussian profile that transformed into a single ring in a Kerr medium [99]. Phase and ray plots revealed that under nonlinear conditions, the super-Gaussian beam was focused into a ring that evolved into a self-consistent G-profile. The ring was unstable to random noise and ultimately collapsed into multiple filaments. The flattened Gaussian profiles induced in the organosiloxane are close approximations of the super-Gaussian profile, which is given by [129]:

$$I(r) = A \exp \left[- \left(\frac{r}{\omega_{SG}} \right)^n \right] \quad (5.4)$$

where A is the maximum value of the function, ω_{SG} , a scale factor and n , the super-Gaussian power, which indicates how rapidly the function decays from its peak to zero. Equation (5.4), which reduces to the Gaussian form when $n = 2$, increases in flatness with n . Plots in Figure 5.3 show the excellent overlap between the super-Gaussian and flattened Gaussian profiles.

The strong correspondence between the two spatial profiles indicates that the origin of ring formation is in fact the same in both Kerr and organosiloxane systems. However, the critical difference is that while single rings can only be obtained with a super-Gaussian beam in Kerr media, they were elicited with an unmodified Gaussian beam in the organosiloxane. In the latter, the flattened refractive index profile does not correspond to the profile of the beam but instead, evolves over time as the refractive index profile induced by the beam reaches saturation.

5.4 Simulations of single ring formation

Two approaches were employed in simulating the formation of a single ring in the organosiloxane. The first one consisted of launching a Gaussian beam in a flattened Gaussian refractive index profile. Then the intensity map resulting from the propagation of the Gaussian beam was exported and converted into a refractive index map.

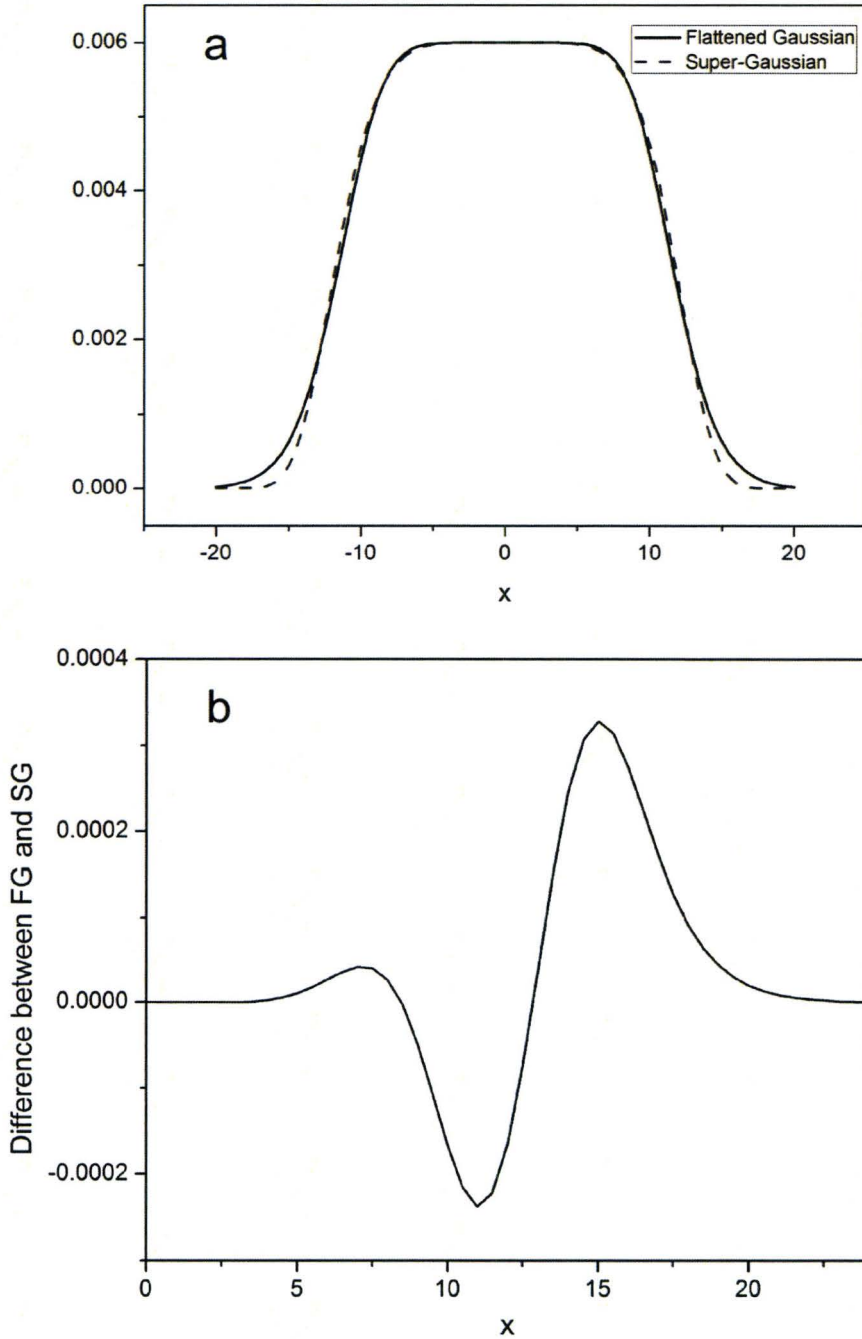


Figure 5.3: a) Comparison between the profiles of a flattened Gaussian beam and a super-Gaussian beam fitted with $A=0.006$, $n=3$ and $\omega_{SG}=12.5$ and b) the difference between both graphs (5 % in the tails) only for one half of the range in x .

This last map served as the new refractive index background for the next simulation step. The second approach is part of the study on simulations of the nonlinear propagation of light at various intensities in the organosiloxane performed in Chapter 6. In there, a Gaussian beam was launched in a uniform refractive index matrix and the resulting intensity map was converted into a map of refractive index which served as the new background refractive index for the next simulation step.

5.4.1 First approach: initial flattened refractive index profile

In the first step of the simulation, a flattened beam was launched into a uniform matrix ($400 \times 6000 \mu\text{m}$) with refractive index 1.47 as seen in Figure 5.4. In the following simulation step, the flat refractive index profile was employed as the background refractive index profile but a Gaussian beam was used as the input beam. The following steps consisted of propagating the input Gaussian beam, exporting values of the 2D intensity profile and converting it into a refractive index profile which served as the background profile for the next simulation. By using the refractive index profile created by the initial input flattened beam, we wanted to recreate the saturation occurring at sufficiently high intensities.

We found that for certain values of the threshold exposure U_0 , the evolution of the input Gaussian beam into a single ring was possible. The U_0 parameter, as explained in Section 6.2 was used to tune the input intensity in the simulations. For instance we can observe in Figure 5.4, the formation of a single-ring for $U_0=0.09$. Here 2D intensity profiles are shown, for step 1, the profile corresponds to the linear propagation of a flattened beam. In step 2, the intensity profile changes drastically, showing at the output face no side bands but a focused beam and mid-way in the propagation distance, at around $4000 \mu\text{m}$ in the propagation direction, two distinctive stripes where most of the intensity is concentrated. As the simulation steps proceed, the fringes reach the exit face of the simulated region (step 26). These fringes represent a single ring intensity profile for a 3D system taking into account an azimuthal symmetry. This is more clearly shown in 1D intensity profiles in Figure 5.5 (For example step 50).

The overall sequence of profiles in the simulations, qualitatively matches the experimental observations as seen in Figure 5.1 up to the formation of the single ring. The formation of the single-ring in Figure 5.1 f of the experiments, can be observed in step 30 of Figures 5.4 and 5.5. Intensity around the ring is greater compared to the initial beam intensity in experiments while in simulations the opposite occurs.

The refractive index profiles corresponding to the simulation steps of Figure 5.4 are presented in Figure 5.6. The initial refractive index profile (step 1) resembles step 1 of the intensity profiles in Figure 5.6, having a conical shape and with traces of the side bands or diffraction rings. We observe that most of the central part reaches the saturation value of $\Delta n_s=0.006$. This indicates that the intensity equivalent to

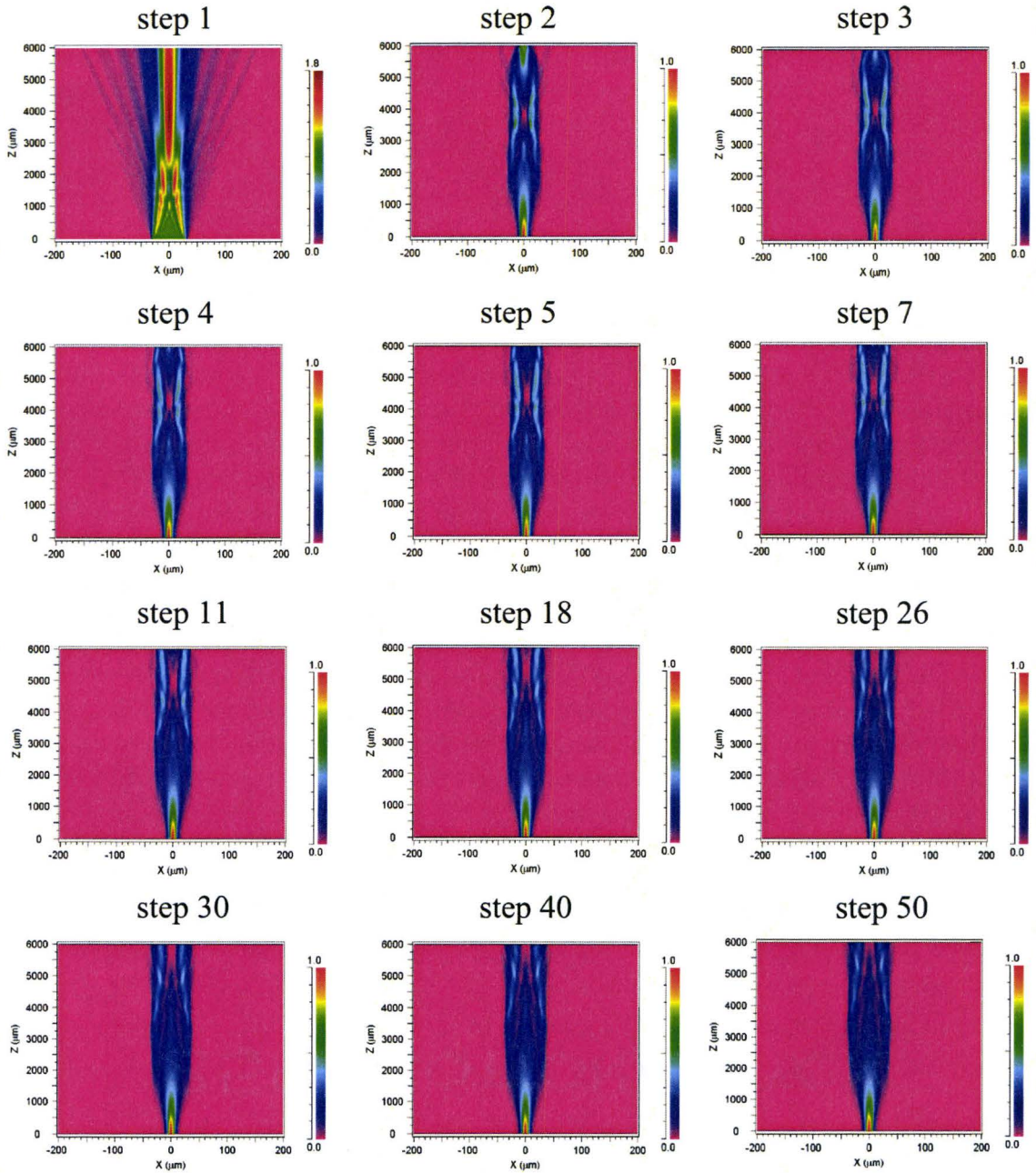


Figure 5.4: 2D intensity profiles resulting from nonlinear simulations of an input Gaussian beam propagated in an initial refractive index profile formed by the linear propagation of a flattened beam and $U_0=0.09$. Over time (steps) the formation of a single-ring is observed. The profiles were normalized to the maximum values of intensity and the display area was zoomed in for better visualization.

Profiles at $z=6.00$ mm

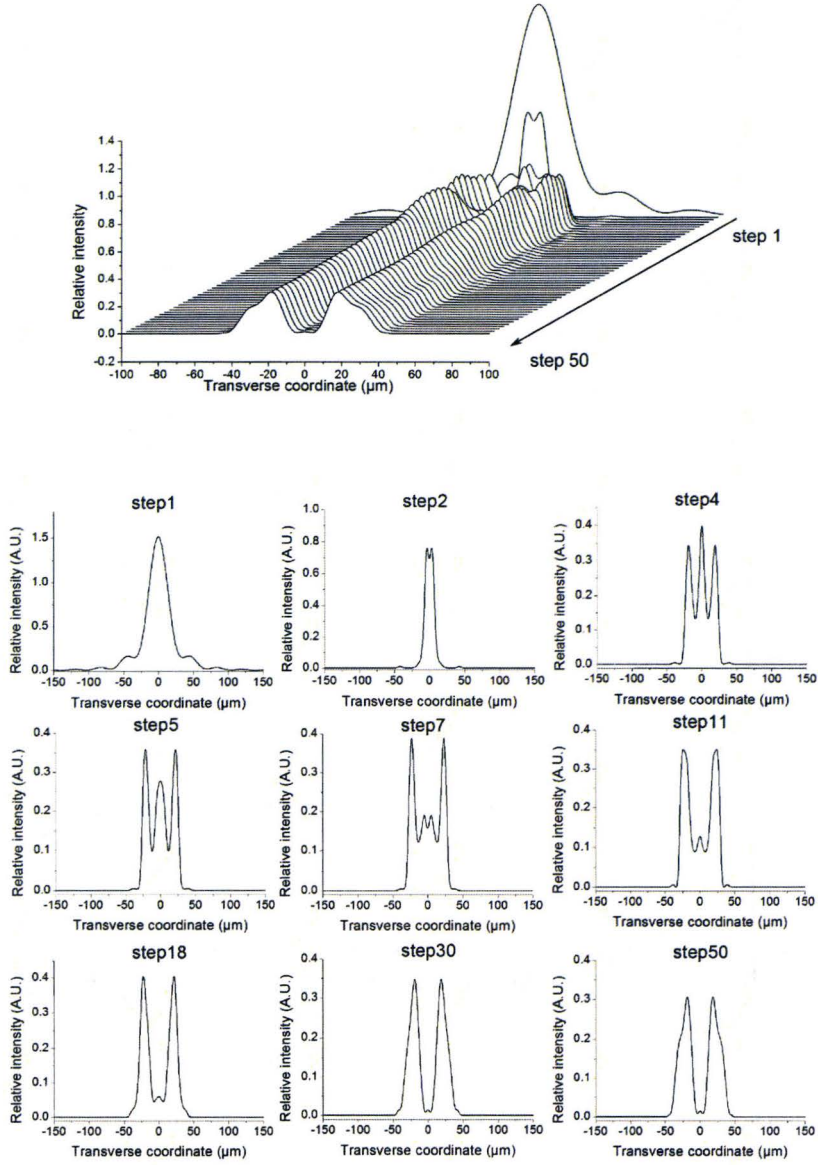


Figure 5.5: Profiles of intensity in the transversal direction x at the output of the medium ($z = 6.00$ mm) for an input Gaussian beam propagated in an initial refractive index profile formed by the linear propagation of a flattened beam and $U_0=0.09$. Corresponding to 50 computational steps (top) obtained in BeampropTM simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit time.

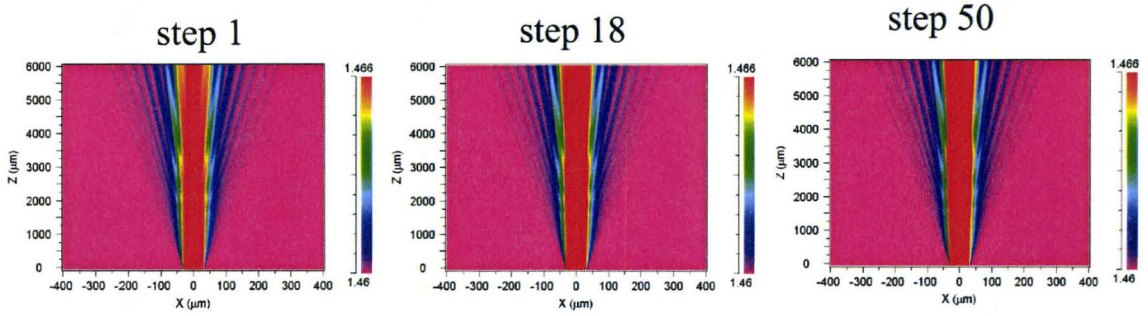


Figure 5.6: 2D refractive index profiles corresponding to the simulations in Figure 5.4 at various computational steps.

$U_0=0.09$ in the simulation is high enough to saturate right from the first step the refractive index in the medium. The refractive index profile for the following computational steps, does not suffer drastic changes. We observe however, further saturation of the refractive index (in step 50) of the central part at the exit face and radially.

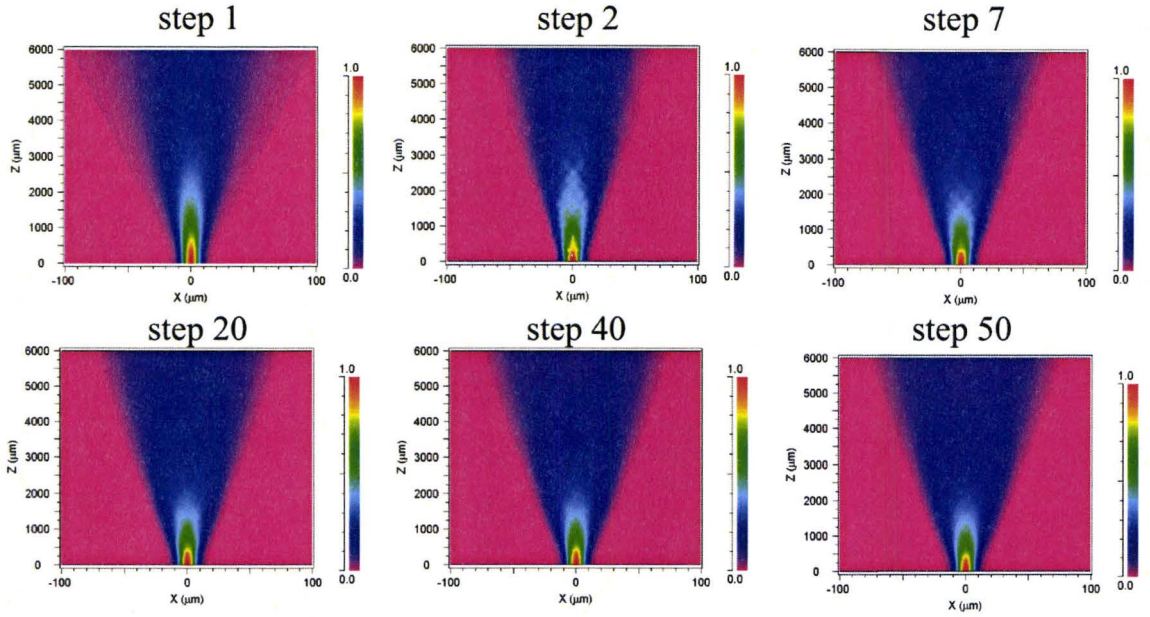
The simulation results show that by having a Gaussian beam launched into a refractive index profile with a flattened-Gaussian shape, a single ring develops at sufficiently high intensities. In the following approach we will examine only the effect of having a Gaussian beam at high intensities.

5.4.2 Second approach: low values of U_0

We also simulated the behaviour of a Gaussian beam in the organosiloxane in an alternate way. A Gaussian beam was directly launched into a uniform matrix (400 X 6000 μm) of refractive index of 1.47 as seen in step 1 in Figure 5.7. The resulting electric field amplitude map was exported and converted into a refractive index map with the external subroutine and then imported in the BeampropTM software for the following steps.

We found that for values of $U_0=0.005$, the Gaussian beam induced a flattened-Gaussian refractive index profile as seen in Figure 5.7b. The evolution of the 2D intensity maps show that initially the Gaussian beam suffers diffraction in step 2 and 7 (Figure 5.7a), followed by a central depression formed at step 40 and 50. More details are observed by plotting the 1D profiles at the output of the medium (Figure 5.8) where few peaks resembling diffraction rings emerge at steps 20 and 30 and at steps 40 and 50, the central dark depression is clearly distinguished having a lobe in each side (Figure 5.8) which indicates the formation of a single ring by using radial symmetry with respect to zero. The contrast in intensity between the central depression and the side lobes however, is too small when compared to experimental results. There, a difference of at least 4-fold exists, whereas in the simulations the

a) Intensity maps



b) Refractive index maps

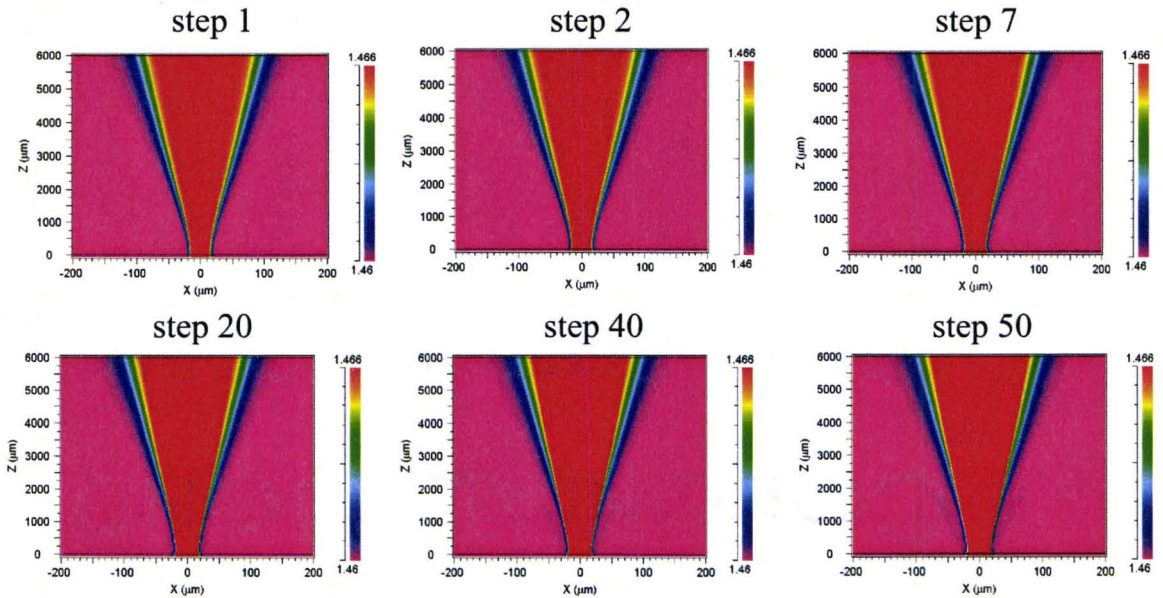


Figure 5.7: a) 2D Intensity maps and b) refractive index maps for $U_0 = 0.005$ for 50 computational steps (top) obtained in Beamprop simulations. Each step represents a relative unit of time.

Profiles at $z=6.00\text{mm}$ for $U_0=0.005$

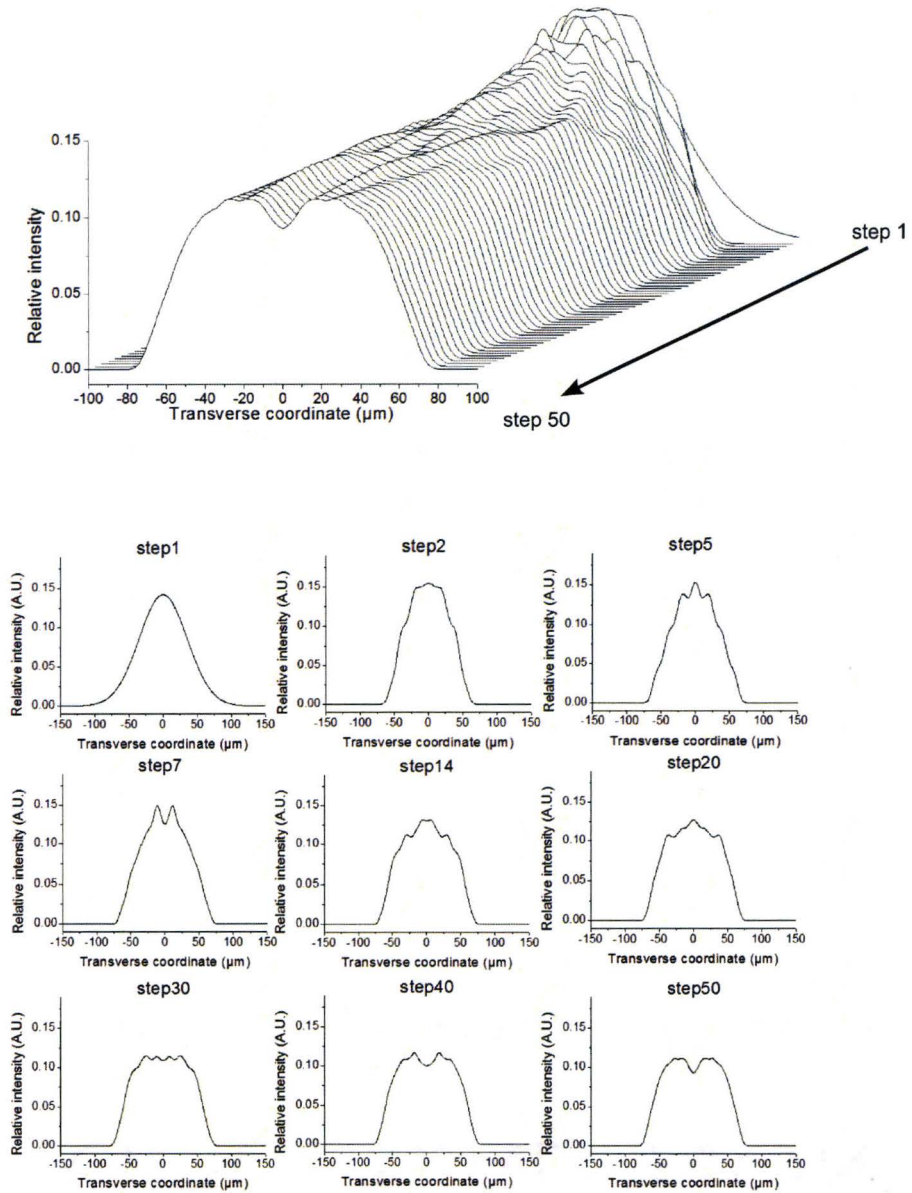


Figure 5.8: Profiles of intensity for $U_0 = 0.005$ in the transversal direction x at the output of the medium ($z = 6.00\text{mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit of time.

contrast is only 1.5 times. The refractive index profile does not change significantly over time, we only observe a slight growth in the saturation area covered by the beam.

Although there is not an exact match between the intensity contrast of the central depression and the side lobes in simulations and the experimental findings, we were able to demonstrate that a Gaussian beam is able to induce a flattened refractive index profile resulting in the formation of a single ring at high enough intensities. Further improvement of simulation parameters and accurate knowledge of physical parameters influencing this process including the maximum refractive index change and critical exposure needed to induce polymerization could help improve the accuracy of simulation results.

5.5 Filamentation of the single-ring

The single ring was unstable to random noise in the organosiloxane and collapsed into multiple filaments (Figure 5.1). We have previously shown that broad, uniform beams propagating in the organosiloxane under similar conditions suffer modulation instability [21]. Weak amplitude perturbations (noise) in the medium that are negligible under linear conditions became amplified, triggering the spontaneous division of the beam into filaments that were randomly positioned in space. In the current study, modulation instability of the single ring led to an azimuthal arrangement of filaments that initially fluctuated in number but rapidly stabilised into 7 filaments. Each filament underwent strong self-trapping as indicated by a 20-fold increase in intensity relative to the initial intensity of the beam and decrease in average filament width ($1/e^2$) from 50 μm at 173 s to 21 μm at 211 s.

Although their spatial positions and number did not change further, careful scrutiny revealed that the intensity distribution within each filament did continue to change. Figure 5.9 traces the behaviour of a single self-trapped filament in the ring.

Until 207 s, the filament retained the tightly focused profile characteristic of a self-trapped beam; this corresponds to the fundamental mode (LP_{01}) supported by a circular waveguide. At 313 s, the intensity distribution within the filament changed into the two lobed profile characteristic of the first order waveguide mode, LP_{11} . The filament reverted back to LP_{01} at 318 s. Such oscillations between LP_{01} and LP_{11} continued until 393 s. Only two oscillations of the beam between LP_{01} and LP_{11} are presented in Figure 5.9.

We showed in Chapter 3 that under certain conditions, a self-trapped beam evolved from single mode to multimode propagation in the organosiloxane. We detected the sequential appearance of up to 5 modes, which underwent oscillations similar to those between the LP_{01} and LP_{11} modes in the filament (Figure 5.9). Self-trapped beams in most other media including Kerr materials and photorefractive crystals induce only a single mode waveguide and propagate as its fundamental mode [117]. Relative to these materials, the maximum refractive index change in the organosiloxane is greater

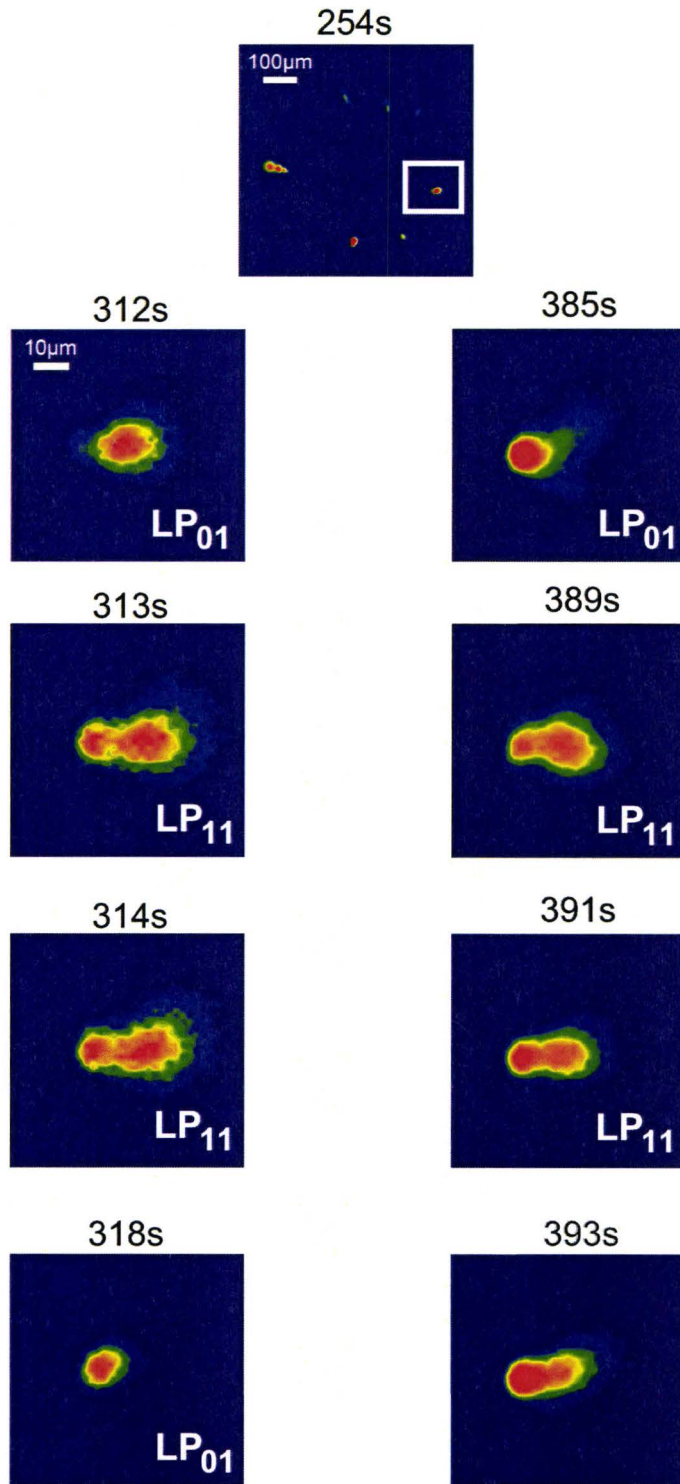


Figure 5.9: 2D intensity profiles showing two sequences of oscillations (columns) where modes appear in a filament for the input intensity of 80 W/cm^2 . At the top, the filament being monitored over time is highlighted with a white square.

by at least 2 orders of magnitude (Section 1.1.4). Here, the beam initially self-traps as the fundamental mode but continues to increase the refractive index of its own waveguide and in this way, sequentially excite high order modes. Theoretical simulations showed that the oscillations of the modes originate from the slowly increasing refractive index of the self-induced waveguide. This causes a continual variation in both the number and propagation constants of modes, which is observed as the complex oscillations between modes at the exit face of the medium. The behaviour of the filaments is similar to a single self-trapped beam. This confirms, as has been proposed by Chiao, Campillo and coworkers [103; 101], that the filaments resulting from modulation instability are a microscale example of self-trapping. In contrast to the self-trapped beam, only one high order mode was observed in the self-trapped filament. As intensity of the beam must be distributed into multiple filaments, the intensity and therefore the refractive index change within a single filament is probably not large enough to support multiple modes. Due to the relatively small dimensions of each self-trapped filament, it was difficult to detect high order modes in all of the repeat experiments. However, we observed them at least twice for each of the intensities studied (*vide infra*).

5.5.1 Single-ring and filamentation dependence on intensity

Single ring formation in the organosiloxane was possible at 6 different intensities including: 27, 40, 64, 80, 95 and 111 W/cm². We experimentally investigated the filamentation and dynamics of the induced single ring at all these intensities.

The overall dynamics of emergence of multiple rings followed by a single ring and then filamentation of the ring were very similar for all intensities as can be seen in Appendix A. However, differences in the diameter of the single-ring induced and number of filaments were observed to be dependent on input intensity.

The first column in Figure 5.10 shows the bright single ring induced at different intensities. The overall trend also shown numerically in Table 5.1, indicates an increase of single ring diameter with increasing intensity for the intensities 40, 64 and 80 W/cm² and then a slight decrease for intensities 95 and 111 W/cm². For the lowest intensity in this range, 27 W/cm², we observed the formation of a single-ring only in one third of the experiments. In those cases, the single-ring diameter was bigger than the next intensity 40 W/cm². When no single ring was formed, the beam directly broke and filamented. This is consistent with previous theoretical and experimental studies in Kerr media, where a single ring results from super-Gaussian beam propagation, an increase in the ring radius with input power has been observed [116; 99].

The number of filaments had an overall increase with input intensity. The maximum number of filaments in which single-rings split for different intensities is observed in Table 5.1. This number starts with two filaments for the intensities 27 and 40

Single-ring formation

Ring filamentation

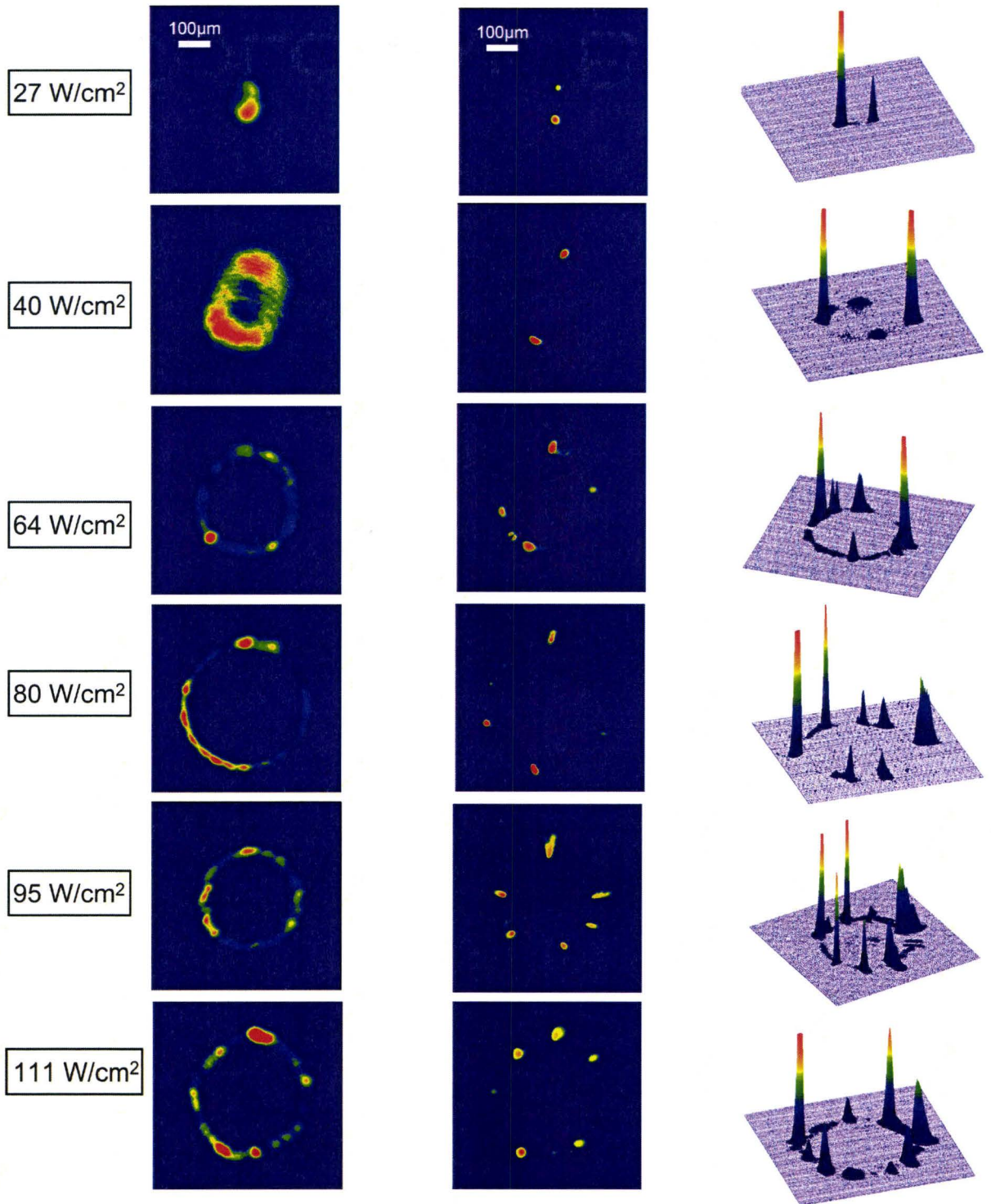


Figure 5.10: 2D intensity profiles of resulting single ring at different input intensities (first column) and the corresponding 2D (second column) and 3D (third column) intensity profiles of filamentation due to instabilities.

W/cm² and it increases to 16 filaments for 64 W/cm², 17 filaments for 80 W/cm² and then it stays at approximately that number for the next two intensities, 95 and 111 W/cm². Although the trend for the maximum and average number of filaments right after starting breakage (see Table 5.2) correlates with the single-ring diameter, i.e. the bigger the ring diameter the greater the number of filaments in which it breaks into, this does not agree for the first intensity of 27 W/cm² in the cases where a single-ring is formed.

Table 5.1: Single-ring diameter and maximum number of filaments

Intensity (W/cm ²)	Ring diameter				Max. # filaments
	Avg. width(μ m)	Stdv. (μ m)	Avg. height(μ m)	Stdv. (μ m)	
27	252	62	317	53	2
40	184	57	248	79	2
64	322	206	409	171	16
80	400	119	442	129	17
95	250	53	290	45	14
111	292	76	327	65	16

Prediction of number of filaments in ring-shaped beams in Kerr media has mostly been performed through azimuthal modulation instability analysis [100]. For optical vortices the number of filaments resulted dependent on power and topological charge (m), which is a measurement of the phase winding [100] and in a saturable Kerr medium, vortex beams resulted in 2m filaments [111]. In rings induced by super-Gaussian beams the number of filaments was proportional to the square-root of the input power [99]. The resulting trend in the organosiloxane is similar to the one found in rings induced by super-Gaussian beams in that the number of filaments increases with input power.

Examining the dynamics of filamentation of the ring, we observed in Table 5.2 that for the first two intensities 27 and 40 W/cm² the number of filaments increased towards the end of the experiment. In contrast for the next four intensities, the filaments mostly fused and their number decreased after the first filamentation. The average filament size was around 30 μ m for the two lowest intensities and around 40 μ m for the remaining intensities. Perhaps at lower intensities, self-trapping of the filaments is more efficient, similar to the self-trapping of the whole beam [21], where at low intensities, narrower waveguides with higher efficiency are formed due to a greater refractive index contrast achieved during self-focusing.

We observed that at the two lowest intensities of 27 and 40 W/cm², the filaments always divided in two in the vertical direction. This correlates with the input beam which is linearly polarized in the same direction, however further investigations should be performed to confirm any polarization dependence on the direction of filamentation. For higher intensities, filaments were more equally spaced at higher intensities compared to lower ones.

Table 5.2: Filamentation parameters

Intensity (power) (W/cm ²)/(μW)	Average # filaments start	Average # filaments end	Average filament size (μm)
27 (85)	2	2.4	31±14
40 (125)	2.4	3.4	31±6
64 (200)	8.7	7	44±12
80 (250)	11.8	8	40±12
95 (300)	9.4	6.6	43±8
111 (350)	11.4	7	40±11

5.6 Permanent structures after filamentation

Refractive index changes resulting from photopolymerization in the organosiloxane are permanent and therefore leave inscribed structures which provide an opportunity to visualize the propagation path of the light after performing the experiments. Figure 5.11 shows the resulting structures for the filamentation experiments for all the intensities studied. We found that the filaments observed in the experiments, induced very thin waveguides that were arranged around a single-ring and their trajectory appears to follow the surface of a cone. In all the cases, the number of thin waveguides matched the number of filaments at the end of the experiments. For example, for the intensity of 27 W/cm² two thin waveguides are observed (Figure 5.11a), in the case of 40 W/cm² four waveguides are observed (Figure 5.11b), since in that experiment the number of filaments changed from two to four towards the end. For higher intensities the number increased according to the final number of filaments observed at the end of the experiment.

5.7 Conclusions

We have shown experimentally the evolution of a Gaussian beam from diffraction rings to a single ring that ultimately suffers filamentation in the organosiloxane for intensities covering from 27 to 111 W/cm². This resulted from having initial intensities large enough to induce a flattening in the refractive index profile. Simulations of the nonlinear propagation of the Gaussian beam confirmed that a flattened refractive index profile leads to the formation of a single ring. Filamentation of the ring was observed as a result of modulation instability. An overall increase in the number of filaments dependent on intensity was found and also the size of the ring was dependent on intensity. And finally, circular arrays of permanently inscribed thin waveguides were observed as a result of the filamentation process along the single ring.

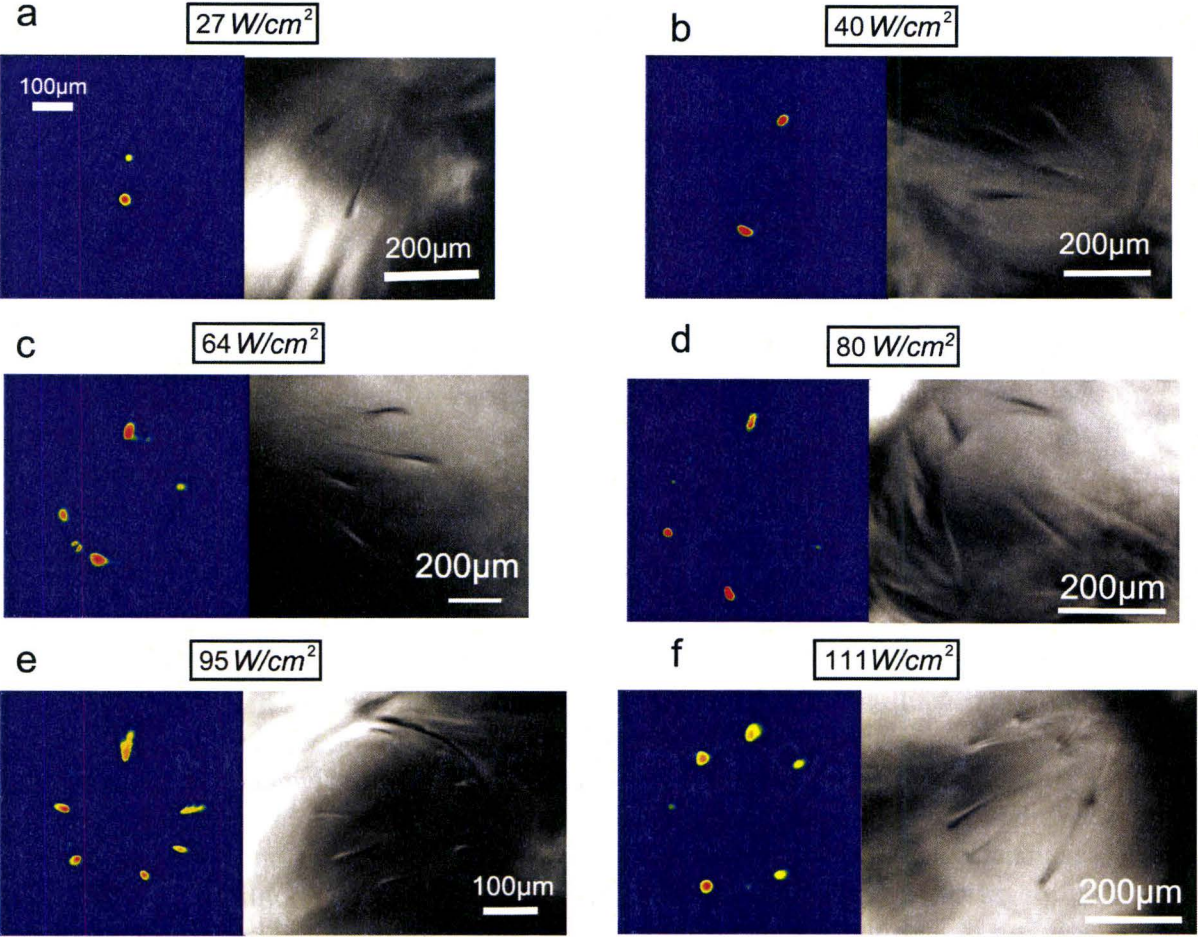


Figure 5.11: 2D intensity profiles of resulting single-ring filamentation at various input powers and their corresponding micrographs of the permanent inscribed structures showing multiple waveguides due to refractive index changes in the organosiloxane

Chapter 6

Simulations of nonlinear propagation at various intensities

The BPM is an approximation of the exact wave equation for monochromatic waves. By using a scalar field assumption, the wave equation reduces to the Helmholtz equation and by using the slowly varying envelope approximation (paraxial approximation) the Helmholtz equation reduces to [118]

$$\frac{\partial u}{\partial z} = \frac{i}{2\bar{k}} \left(\frac{\partial^2 u}{\partial^2 x^2} + \frac{\partial^2 u}{\partial^2 y^2} + (k^2 - \bar{k}^2)u \right) \quad (6.1)$$

having

$$\phi(x, y, z) = u(x, y, z)e^{i\bar{k}z} \quad (6.2)$$

where $u(x, y, z)$ is the slowly varying field, $\bar{k}$ is a constant number that represents the average phase variation of the field $\phi(x, y, z)$, k is the wave number and x and y are the transverse coordinates and z is the propagation coordinate. We performed 2-D simulations in BeamPROPTM, which is a simplification of Equation (6.1) where the field is considered to be extended infinitely and the y dependence is omitted. This can be justified by the radial symmetry of refractive index changes in the isotropic organosiloxane.

Numerical simulations of refractive index profiles induced at different intensity regimes are critical since we found in our experimental work that they determine the type of nonlinear phenomenon observed. We deduced that in the low intensity regime (see Chapter 3), a Gaussian refractive index profile was induced in order to observe self-trapping. In the mid-intensity regime, a Gaussian index profile with greater refractive index change values compared to the low intensity index profile was needed for the emergence of diffraction rings. At the high intensity regime, a flat top refractive index profile is induced, which prevented self-trapping and allowed for the formation of a single ring and filamentation in the organosiloxane.

6.2 Equivalence of intensity and U_0 in simulations

Simulations over a wide range of intensities were performed using the software

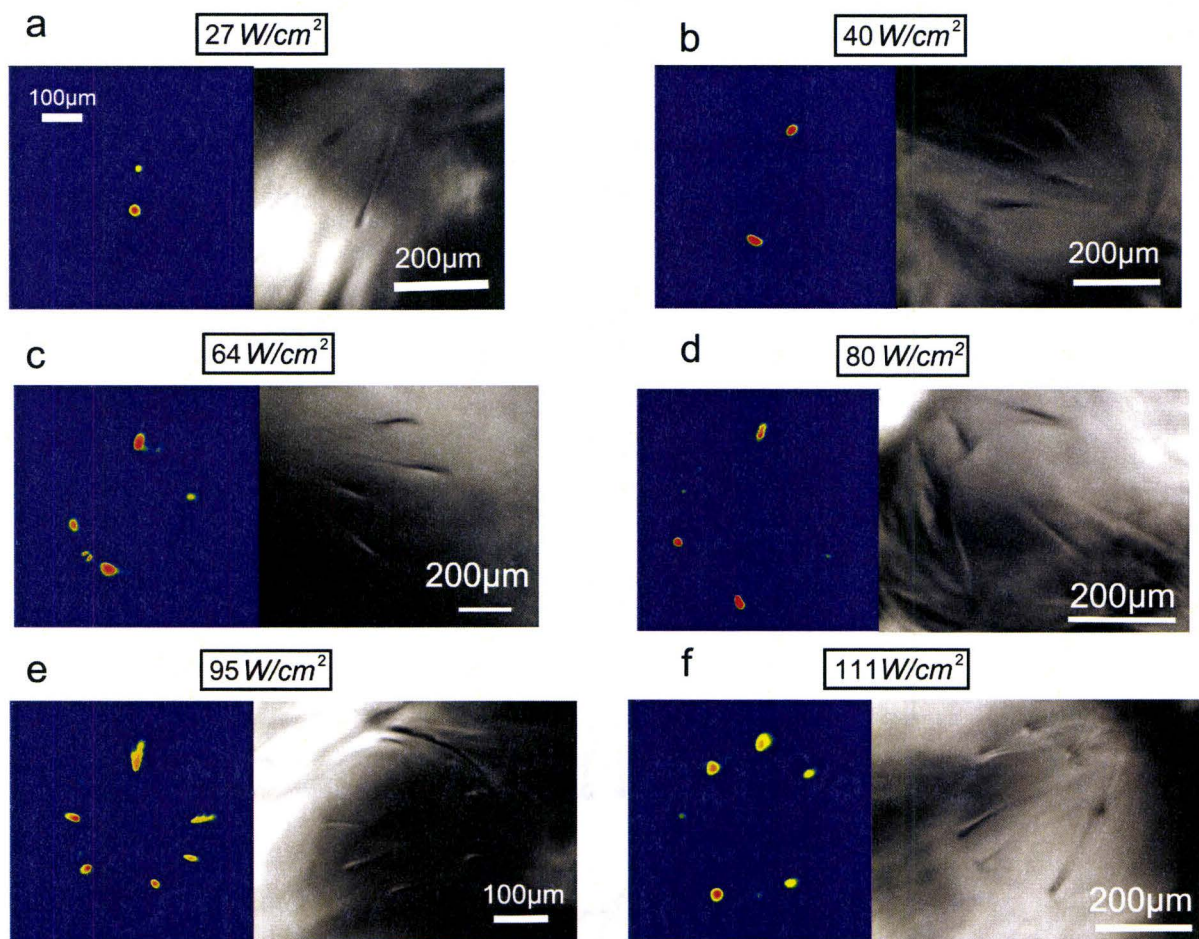


Figure 5.11: 2D intensity profiles of resulting single-ring filamentation at various input powers and their corresponding micrographs of the permanent inscribed structures showing multiple waveguides due to refractive index changes in the organosiloxane

Chapter 6

Simulations of nonlinear propagation at various intensities

In previous chapters, we presented experimental studies of laser propagation in a photopolymer covering a wide range of intensities. A variety of phenomena was observed at different intensities including self-trapping, excitation of high order modes, diffraction rings, single ring formation and filamentation.

Previous theoretical studies have examined self-trapping in photopolymers through two-dimensional numerical simulations using the beam propagation method and the measured index evolution [20], and also by using the paraxial wave equation with one transverse dimension and updates in the refractive index change [130; 131; 35]. Wentzel-Kramers-Brillouin analysis has been employed to understand the modal properties of the waveguides formed in photosensitive materials during self-trapping [63]. Theoretical studies on diffraction ring phenomena have been mainly realized for thin media (thickness $\leq$ Rayleigh range) by addition of the phase shift induced by refractive index changes to the output amplitude of the electric field. The far-field intensity distribution has then been obtained by applying the Fraunhofer approximation of the Fresnel-Kirchhoff diffraction formula to the electric field amplitude [85]. For long pathlengths (thickness $>$ Rayleigh range), diffraction ring theory has been developed for sodium vapour by employing a paraxial approximation of Maxwell equation for a homogeneously broadened two-level system [81]. Photorefractive crystal shock waves (closely related to diffraction rings) have been modelled in the context of fluid dynamics using the Euler-like fluid equations [80]. Single-ring formation during nonlinear light propagation has been modelled in Kerr media through the nonlinear Schrödinger equation and its filamentation through an azimuthal modulational instability analysis [99]. Filamentation of photopolymers has been modelled by adding small transverse and spatial perturbations to plane waves [105] based on the modulation instability theory from Bespalov and Talanov [96].

We performed numerical simulations of a c.w. laser nonlinear propagation in the organosiloxane in two dimensions (2D) since previous theoretical work on nonlinear phenomena has not modelled all the features and trends observed in our experimental work. For instance, no theoretical studies have been performed on intensity dependent self-focusing and self-trapping dynamics. Diffraction ring emergence has not yet been modelled in photopolymers and theory for long pathlengths has been limited to very different systems [81]. Single ring formation has only been modelled for the non-saturable Kerr medium where the nonlinear mechanism is fundamentally differ-

ent. Overall, there is a lack of systematic numerical studies of the intensity dependent nonlinear light propagation and corresponding refractive index changes in photopolymers. In this study, we address this problem by performing numerical simulations over a wide range of intensities in a photopolymer. We simulated most of the features observed in our experimental work including temporal dynamics of self-trapping in the low intensity regime, modal correlation with oscillations during self-trapping, the emergence of diffraction ring and single ring formation.

In the following Sections, we present first an overview of the methodology employed to perform simulations using BeamPROPTM software and an external subroutine (Appendix B). This is followed by the equivalence between tuning the value of critical exposure U_0 and varying the intensity. Results of the simulated dynamics of self-trapping at the low intensity regime are then presented including self-trapping trends, dynamics of modal evolution and dependence of self-trapping with intensity. Simulations in the mid-intensity regime are presented with the emergence of diffraction rings. At higher intensities, we simulated the formation of a single ring. Finally simulations at very high intensities are presented. We will show that this approach models the different phenomena observed in our experimental studies including self-trapping, oscillations accompanied with high-order modes, diffraction rings and single-ring formation. Filamentation of the beam was not simulated because this requires introduction of random noise in the system which increases the modeling complexity.

6.1 Nonlinear propagation in organosiloxane using BeamPROPTM

We modelled nonlinear propagation of a Gaussian beam in the organosiloxane through an iterative process using the beam propagation method (BPM) combined with the calculation of refractive index changes in the medium. The BPM was used with the aid of the software package BeamPROPTM (RSoft Design Group, Inc) and the refractive index calculations were performed by using an external subroutine (Appendix B). Specifically, a Gaussian beam was launched in a uniform refractive index medium, its propagation under linear conditions was calculated and a map of the resulting electric field amplitude along the propagation length was obtained. Then, using Equation (6.3) in the external subroutine, refractive index changes were calculated based on the electric field amplitude map and those changes were input in the BeamPROPTM software where the propagation of the Gaussian beam through this modified medium was modelled again. This process was iteratively performed over many steps. This approach is equivalent to nonlinear propagation in the photopolymer; by updating the new changes in refractive index in the medium we take into account the nonlinearity of the system. The results obtained by using this approach are consistent with numerical results on self-trapping by Monro and coworkers [35; 124]. For details of the simulation procedure see Section 2.5.1.

The BPM is an approximation of the exact wave equation for monochromatic waves. By using a scalar field assumption, the wave equation reduces to the Helmholtz equation and by using the slowly varying envelope approximation (paraxial approximation) the Helmholtz equation reduces to [118]

$$\frac{\partial u}{\partial z} = \frac{i}{2\bar{k}} \left(\frac{\partial^2 u}{\partial^2 x^2} + \frac{\partial^2 u}{\partial^2 y^2} + (k^2 - \bar{k}^2)u \right) \quad (6.1)$$

having

$$\phi(x, y, z) = u(x, y, z)e^{i\bar{k}z} \quad (6.2)$$

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Numerical simulations of refractive index profiles induced at different intensity regimes are critical since we found in our experimental work that they determine the type of nonlinear phenomenon observed. We deduced that in the low intensity regime (see Chapter 3), a Gaussian refractive index profile was induced in order to observe self-trapping. In the mid-intensity regime, a Gaussian index profile with greater refractive index change values compared to the low intensity index profile was needed for the emergence of diffraction rings. At the high intensity regime, a flat top refractive index profile is induced, which prevented self-trapping and allowed for the formation of a single ring and filamentation in the organosiloxane.

6.2 Equivalence of intensity and U_0 in simulations

Simulations over a wide range of intensities were performed using the software BeamPROPTM and an external subroutine (Appendix B). However, BeamPROPTM does not allow for changes to the input beam intensity, it only allows for an input beam with a unitary electric field amplitude. To overcome this limitation, we tuned the parameter critical exposure U_0 in the formula for the refractive index change of the external subroutine, which was equivalent to changing intensity.

To understand the equivalence between U_0 and incident intensity, we can first examine the formula employed to convert electric field amplitude maps to refractive index maps. It corresponds to the formula developed by Kewitsch and coworkers [39]:

$$\Delta n(x, y, z, t) = \Delta n_s \left\{ 1 - \exp \left[-\frac{1}{U_0} \int_0^{t-\tau} |E(t)|^2 dt \right] \right\} \quad (6.3)$$

where Δn_s is the maximum refractive index change, U_0 is the critical exposure required to initiate polymerization, τ is the monomer radical lifetime and $E(t)$ is the amplitude of the electric field.

If we were to tune the intensity ($|E(t)|^2$) and fix the other parameters (Δn_s and U_0), we would obtain a set of refractive index change curves which saturate at different times as observed in Figure 6.1 A.

In this case we fixed the values of U_0 and Δn_s to 1 J/cm² and 0.006 [24] respectively. In Figure 6.1 B, we fixed the value of intensity at 1 W/cm² and of Δn_s at 0.006 and vary U_0 , obtaining equivalent refractive index curves over exposure time. To obtain the equivalent U_0 for a particular intensity, we calculated the ratio of $\int_0^{t-\tau} |E(t)|^2 dt^2 / U_0$ for each of the values in Figure 6.1 A. In that case, we took the numerator as the intensity over one second, which corresponds to radiant exposure. For example, for the intensity 12732 W/cm², the numerator would correspond to 12732 Ws/cm² or J/cm² and the denominator to $U_0=1$ J/cm², therefore the ratio $\int_0^{t-\tau} |E(t)|^2 dt^2 / U_0$ is 12732. To find the equivalent U_0 , we just solve for the equation and having $\int_0^{t-\tau} |E(t)|^2 dt^2 = 1$ W/cm² · 1 s = 1 J/cm², we obtain $U_0 = 1 \text{ J/cm}^2 / 12732 = 0.00008 \text{ J/cm}^2$. In order to obtain a direct relation between the experimental intensities employed and the simulated values U_0 , the physical value of critical exposure in the system is required.

We can understand the equivalence between intensity and the parameter U_0 since the value of U_0 is the critical exposure to initiate polymerization, therefore the smaller this value is, the faster changes in refractive index will occur and *viceversa*. Therefore, small values of U_0 represent high intensities and larger values of U_0 represent lower intensities.

To further test the equivalence between varying the parameter U_0 and tuning intensity, we plotted the refractive index profiles induced by Gaussian beams of varying intensity (fix U_0) and of varying U_0 (and fix intensity) as observed in Figure 6.2. In Figure 6.2A1, input Gaussian beams with varying intensities are shown and in Figure 6.2A2, the corresponding refractive index induced. Figure 6.2B1 in contrast shows only a single Gaussian beam intensity and Figure 6.2B2 the induced refractive index profiles at values of U_0 that are equivalent to intensities in Figure 6.2A1. We observed that the exact same refractive index profiles are induced in both cases. This proves that changing U_0 values achieves a similar effect on refractive index changes as tuning intensity.

To summarize our approach, we will simulate the intensity dependent nonlinear propagation of light in the organosiloxane by iteratively propagating a Gaussian beam using BeampropTM combined with calculations of refractive index changes. To vary the intensity, the parameter U_0 will be tuned in Equation (6.3) of the external subroutine. A medium pathlength of 6.00 mm will be employed and a 20 μm diameter Gaussian beam will be launched at a wavelength of 532 nm in the central part of a 2D block (800 μm X 6.00 mm) with uniform refractive index. These values correspond to

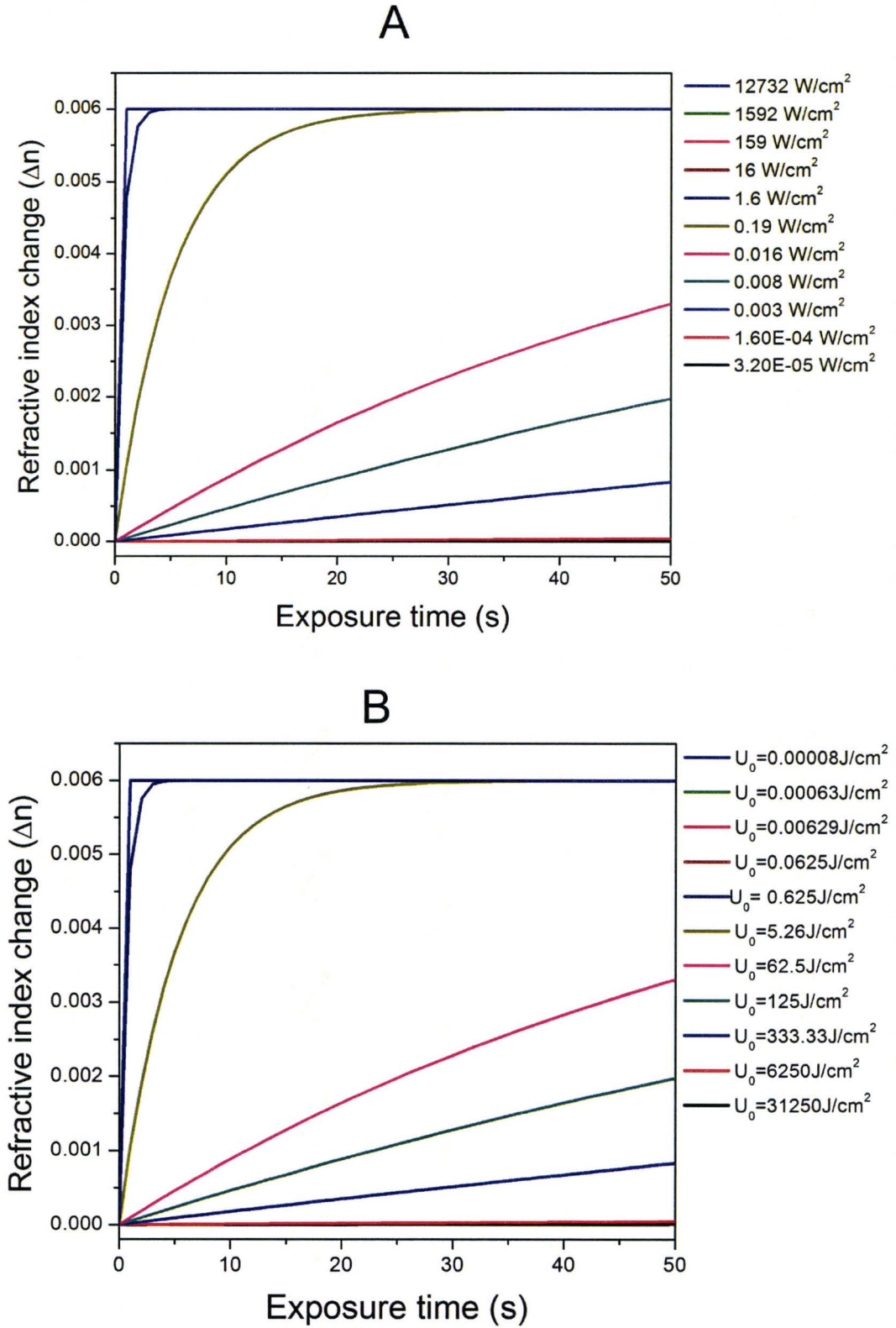


Figure 6.1: Graphs of refractive index change with respect to exposure time based on Equation (6.3) for A) varying intensity and for B) varying U_0 .

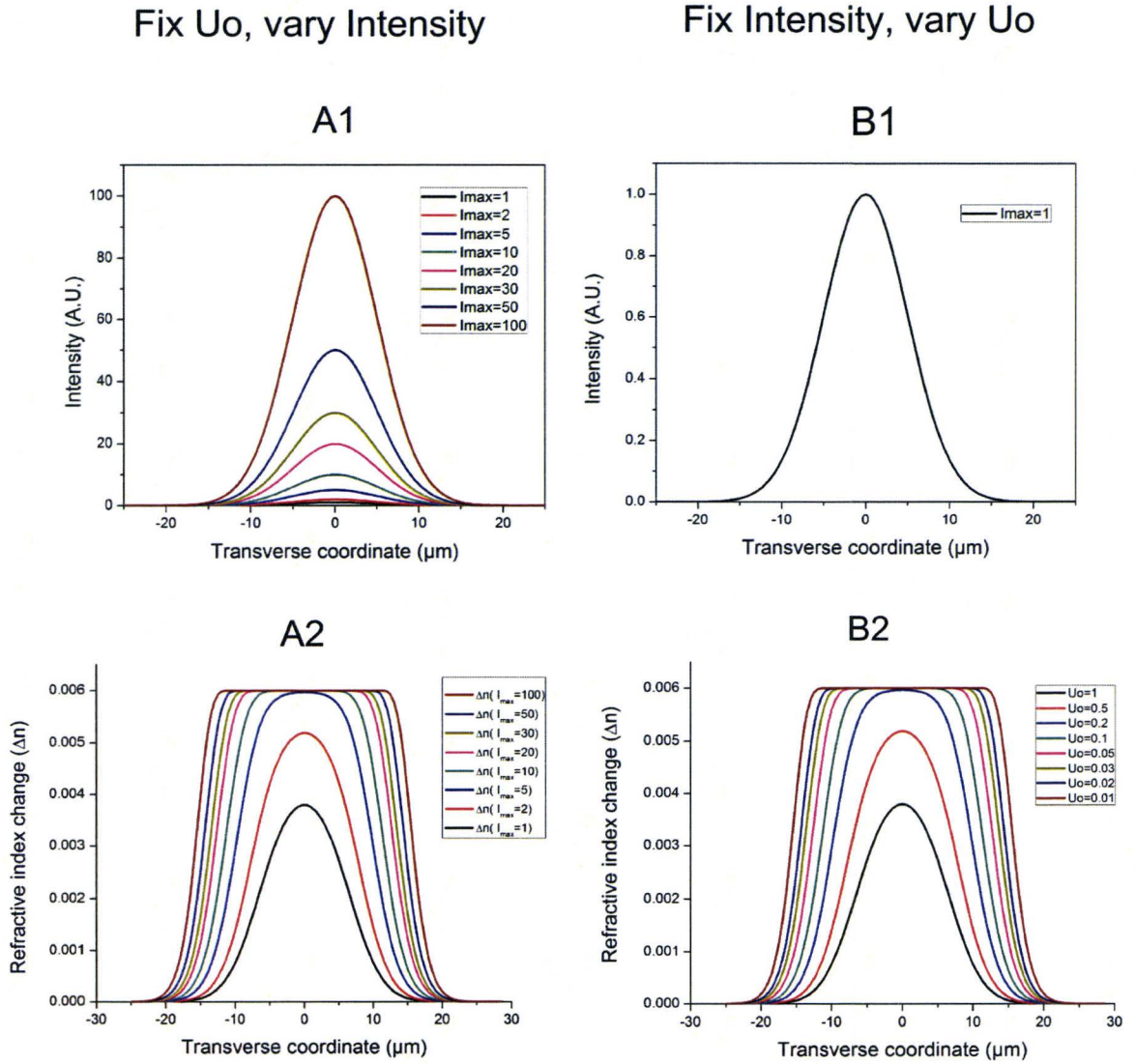


Figure 6.2: Equivalence of refractive index profiles induced by A) varying intensity and B) varying critical exposure U_0 . Figure A1 indicates the different input intensities and Figure A2 the corresponding refractive index profiles. Figure B1 has a single input intensity and Figure B2 indicates the resulting refractive index profiles for various values of U_0 .

experimental values except for the width of the medium ($800\ \mu\text{m}$) which was reduced to optimize computation time. In numerical simulations we will be probing the three intensity regimes that were found in our experimental studies.

6.3 Dynamics of self-trapping in the low intensity regime

In the low intensity regime, we performed simulations for values of $U_0=500, 100, 50$ and 10 . Dynamics of self-trapping were modelled including changes in the intensity and refractive index profiles over time. One-dimensional transverse profiles of intensity along z and at the output of the medium ($z=6.00\text{mm}$) were obtained over time giving information in the modal composition. Oscillations of the beam intensity at the output of the medium accompanied with the observation of high-order modes were also confirmed using simulations. Obtained simulated trends for the intensity dependent parameters for self-trapping agreed with the experimental ones, showing that an optimum intensity exists at which the beam self-traps more efficiently.

6.3.1 Self-trapping

For the lowest intensity in simulations, $U_0=500$, Figures 6.3 and 6.4 show the resulting intensity and refractive index maps, respectively, for computation steps covering from step 1 to step 70. Each step represents a unit of time in simulations. The initial intensity profile (step 1) in Figure 6.3 shows the diffraction of the input Gaussian beam through the medium. The corresponding refractive index profile shows that in the first step (Figure 6.4) the beam is only able to polymerize areas of high intensity forming a bullet-like structure.

Over time, we observe the beam overcoming diffraction indicated by the narrowing of its width (steps 4 to 29 in Figure 6.3). The intensity maximum observed at the input face $z=0.00\ \text{mm}$ in step 1, moves away along the propagation axis in the following steps, reaching close to $z=50\ \mu\text{m}$ in step 29. The translation of the maximum (called also primary eye) was explained to occur when the refractive index change in the material is large enough to overcome the initial beam diffraction and therefore it was considered as a precursor to waveguide formation [35]. At step 25 (Figure 6.3), the intensity maximum has moved away from the input face and the beam has narrowed significantly counteracting diffraction. At the same time, we observed that the refractive index profile at step 25 (Figure 6.4) started to form a channel waveguide and at step 70 a thin and uniform waveguide had formed.

Formation of multiple intensity maxima is observed along the propagation axis in Figure 6.3. Three, four and nine intensity maxima are formed in steps 34, 50 and 70, respectively for $U_0=500$. The appearance of intensity maxima has been explained as a result of mode beating [63]. Our results are generally consistent with previous

$U_0=500$ Intensity maps

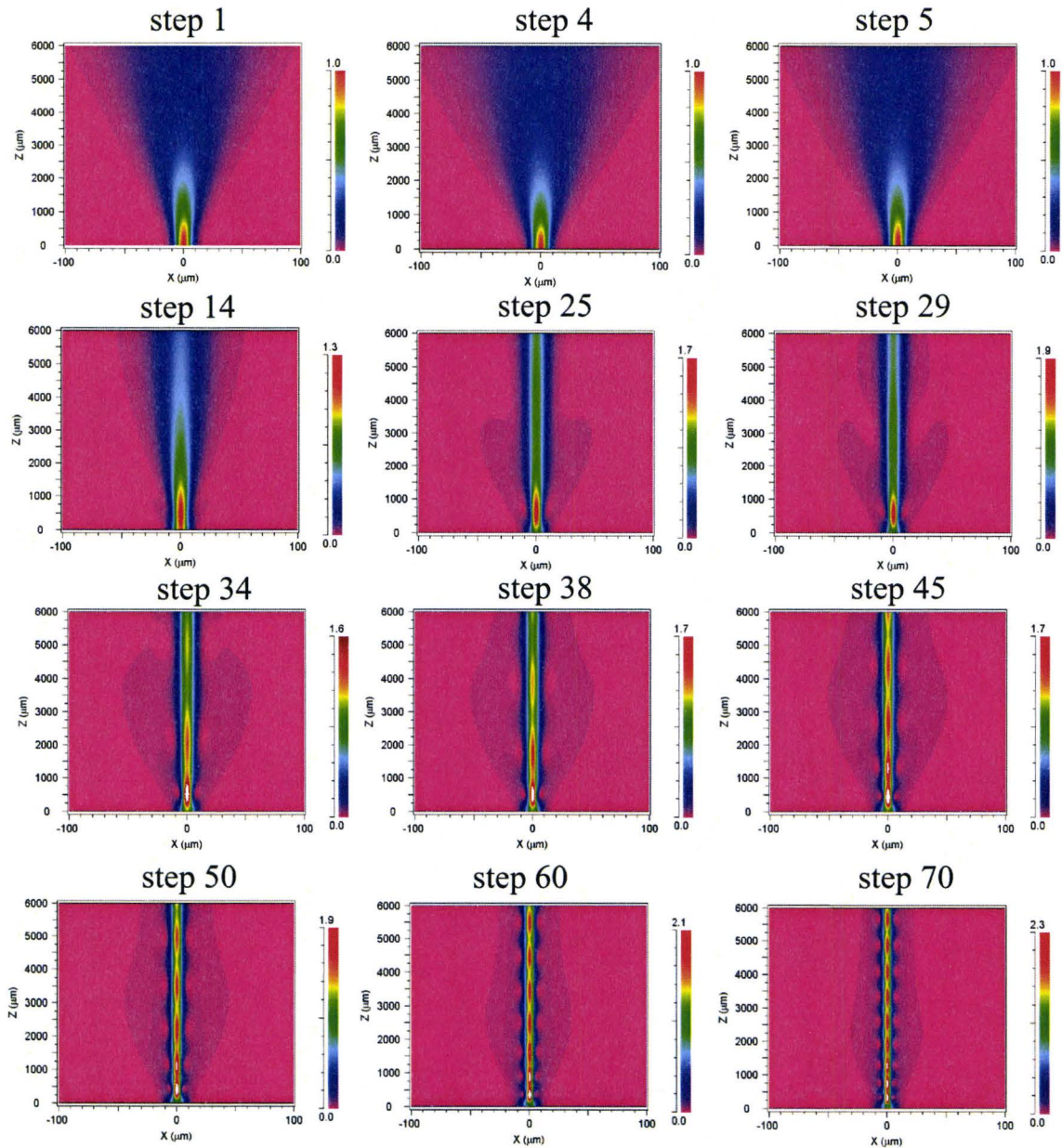


Figure 6.3: 2D Simulation results in Beamprop showing the intensity profiles of a propagating Gaussian beam with $U_0 = 500$. The propagation coordinate is indicated with z and the transverse coordinate with x

$U_0=500$ Refractive index maps

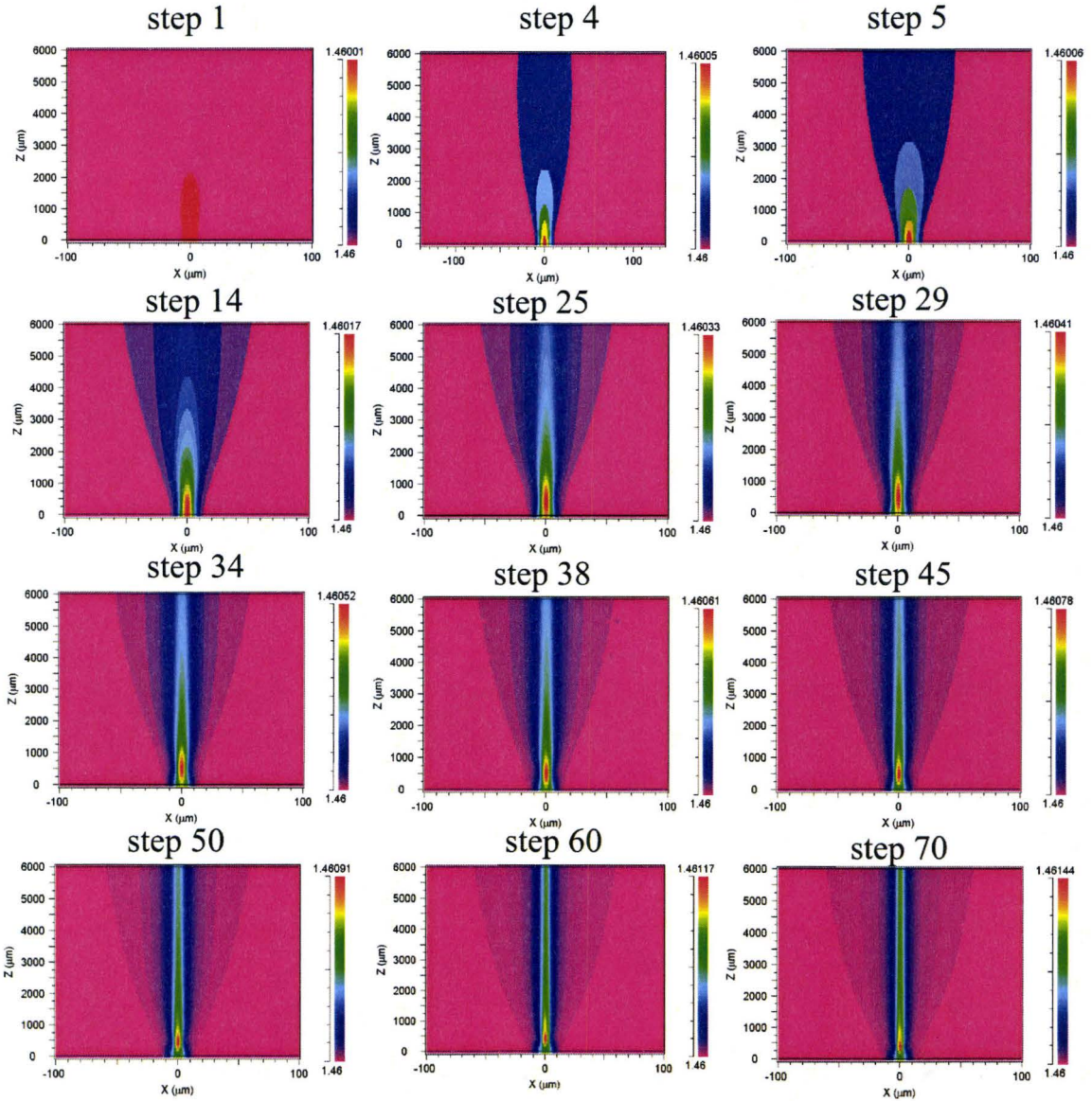


Figure 6.4: 2D Simulation results in Beamprop showing the refractive index profiles of a propagating Gaussian beam with $U_0 = 500$. The propagation coordinate is indicated with z and the transverse coordinate with x

simulations developed by Monro and coworkers [35] however details of the dynamics of mode evolution has not been previously modelled. This will be addressed in the following Sections taking into account a larger magnitude of Δn compared to previous simulations.

6.3.2 Dynamics of modal evolution

The intensity profile along the propagation axis z did not remain constant during simulations, which resulted from the formation of intensity maxima during self-trapping. These maxima were more intense closer to the entrance face and decreased as the beam propagated through z as seen in Figure 6.5. Intensity profiles were narrower at locations of maximum intensity and wider at locations of minimum intensity. More detail can be found in Figure 6.6a, where the intensity profiles for the first oscillation along z of step 50 are plotted from $z=1100\ \mu\text{m}$ and $z=2100\ \mu\text{m}$. These correspond to locations of intensity maxima. The intensity profile at the first maximum presents two side lobes next to the peak, a similar profile is repeated in the next maximum but broadened. At the location of minimum intensity, $z=1600\ \mu\text{m}$, the intensity profile became wider and intensity seemed to be transferred to the sides of the peak. A similar behaviour was observed at the last oscillation of step 50 from $z=3400\ \mu\text{m}$ to $z=4800\ \mu\text{m}$ shown in Figure 6.6b. Here the difference was that all the intensity profiles had broadened significantly compared to the ones shown in the first oscillation. Intensity maxima in step 50 were found to be positioned aperiodically at increasing distances along z , starting from around $1000\ \mu\text{m}$ in the first oscillation and ending with a separation of $1500\ \mu\text{m}$ in the last oscillation.

Overtime, we observed the appearance of more intensity maxima as shown in Figure 6.7. Here, intensity maxima for a later step (step 70) are shown. A behaviour similar to the one in step 50 occurred with few differences. The side lobes of the intensity maxima ($z=700\ \mu\text{m}$ and $z=1300\ \mu\text{m}$) for the first oscillation (Figure 6.8a) were higher in intensity compared to the first oscillation of step 50. Intensity profiles for the last oscillation in step 70 (Figure 6.8b), had a width that was narrow compared to the last oscillation for step 50 (Figure 6.6b). Also there was more similarity in intensity profiles between first and last oscillations in step 70 compared to the ones in step 50. The position of the intensity maxima at step 70 became more periodic along z and as more intensity maxima emerged, the distance between them was smaller. The distance between the first maxima was around $400\ \mu\text{m}$, for the second and third $600\ \mu\text{m}$ and the distance between the last maxima was around $700\ \mu\text{m}$.

These simulations, confirmed that not only the fundamental mode propagated along z during self-trapping but also high order modes. The modal composition was observed to vary along z for step 50 (Figures 6.5). This was indicated by the different intensity profiles observed during oscillations and in the propagation distance. In the first oscillation (Figures 6.6a), high intensity side lobes occurred at locations of intensity maxima (at $z=1100\ \mu\text{m}$). This can be explained as a superposition

Profiles along z for step 50, $U_0=500$

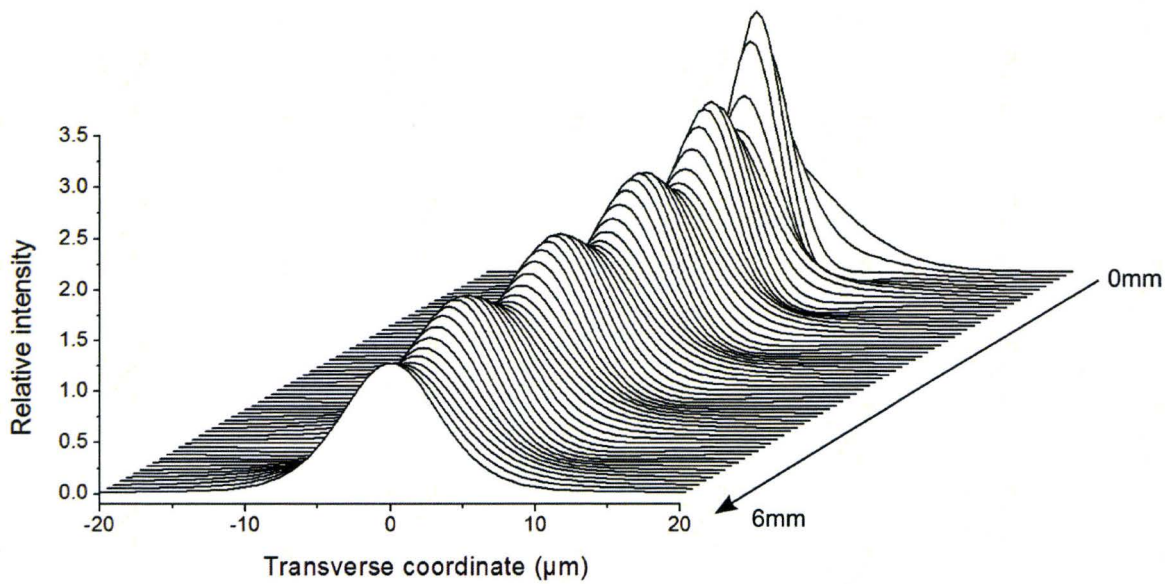


Figure 6.5: Simulated intensity profiles along the propagation axis z for step 50 of a propagating Gaussian beam with $U_0 = 500$.

Profiles of step 50 at various "z" for $U_0=500$

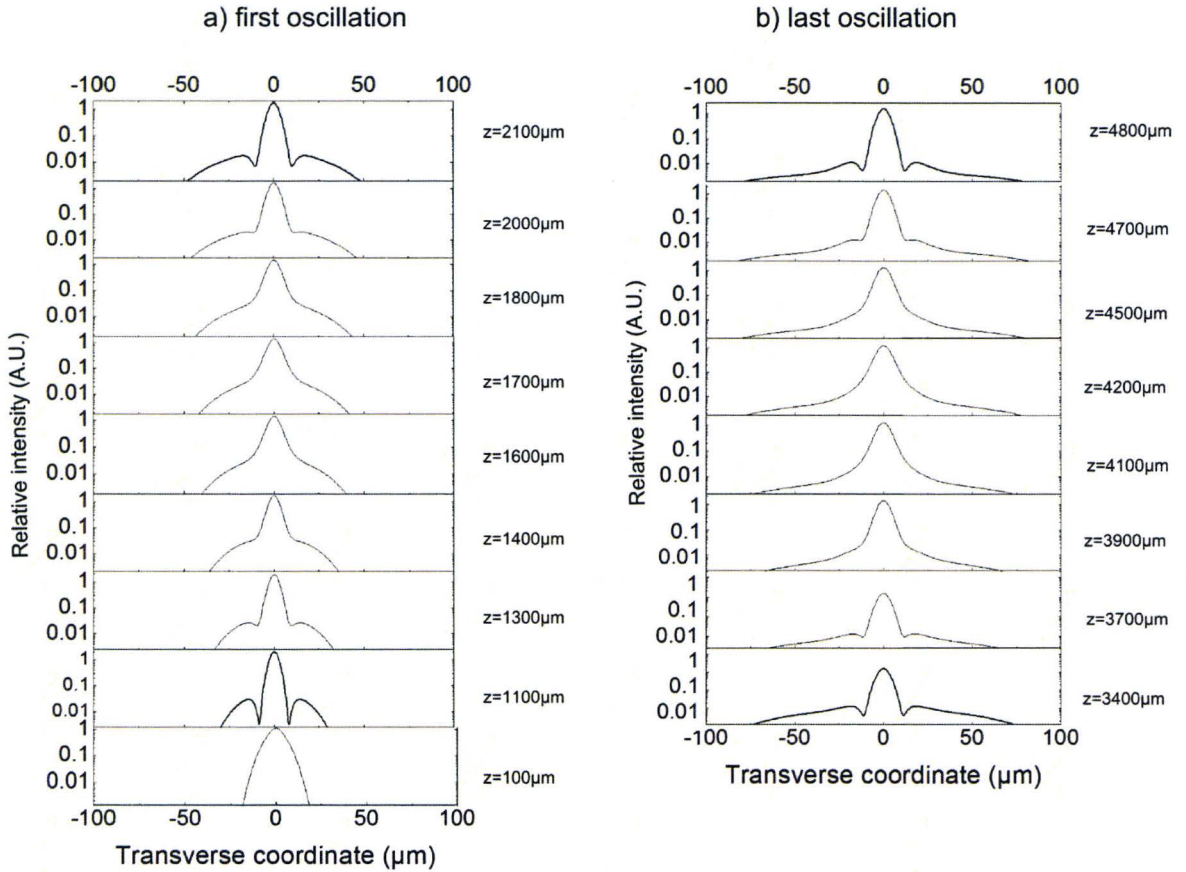


Figure 6.6: Simulated intensity profiles for the a) first and b) last oscillations along the propagation axis z for step 50 of a propagating Gaussian beam with $U_0 = 500$. In bold are indicated the profiles for the intensity maxima in the oscillation. The relative intensity is on a logarithmic scale for clarity.

Profiles along z for step 70, $U_0=500$

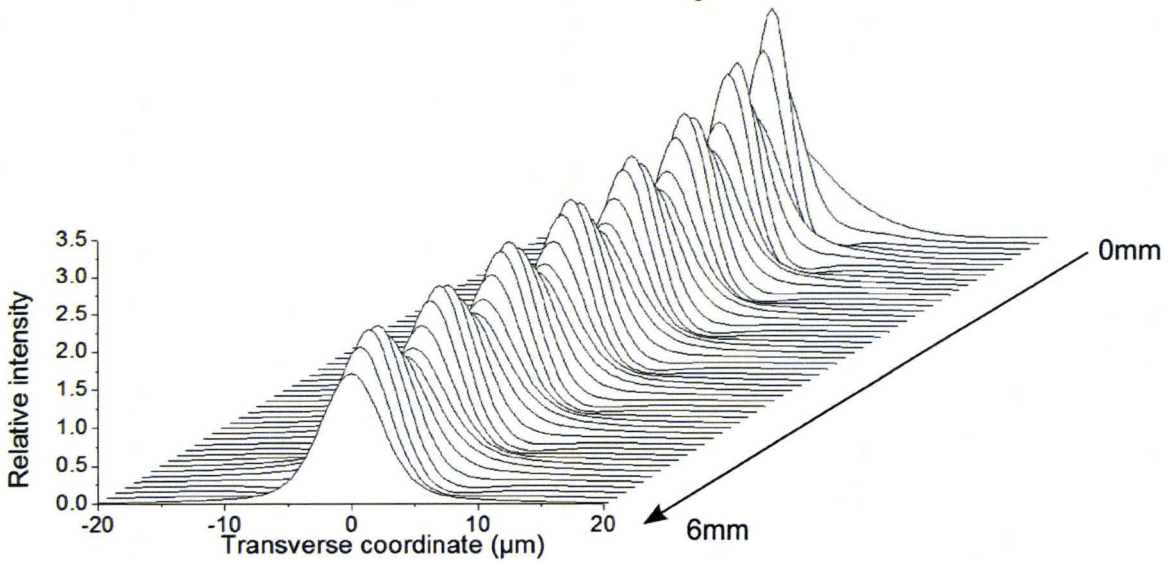


Figure 6.7: Simulated intensity profiles along the propagation axis z for step 50 of a propagating Gaussian beam with $U_0 = 500$.

between modes, which could represent the superpositions of the fundamental mode with either LP_{02} , LP_{11} or LP_{21} . At intensity minima, the fundamental mode was either very weakly guided or not guided at all and a superposition of high order mode took place, as the peak intensity decreased and side lobes intensity also increased and broadened. At the minimum, $z=1600 \mu\text{m}$, (Figure 6.6a) the superposition of LP_{02} with either LP_{11} or LP_{21} occurred. We observed that in the last oscillation for step 50 (Figures 6.6b) there was a change in intensity profiles compared to the first oscillation and therefore a likely change in modal composition. This could be the result of the variation in the refractive index value and profile along z during self-trapping (Figure 6.4). As the index profile also varied over time, changes were observed in intensity profiles and therefore changes in modal composition in step 70 (Figure 6.8) compared to step 50. The greater similarity between the first and last oscillations of step 70 compared to the ones in step 50 can also be explained with the evolution of the index profile along z . As the induced waveguide became more uniform in z (Figure 6.4), the propagation constants of the excited modes also became approximately constant along z and the modal composition did not vary as much for step 70. This also explains the increasing spatial periodicity of the intensity maxima at step 70, as the beat length of the modes would become more constant.

The overall behaviour of the narrowing of the beam, the formation of a thin waveguide and oscillations in the simulations is in agreement with our experimental observations. We observed that intensity maxima along z correlated with the existence of multiple modes in the induced waveguide during self-trapping. However, in order to compare directly the experimental intensity profiles of the beam with the

Profiles of step 70 at various "z" for $U_0=500$

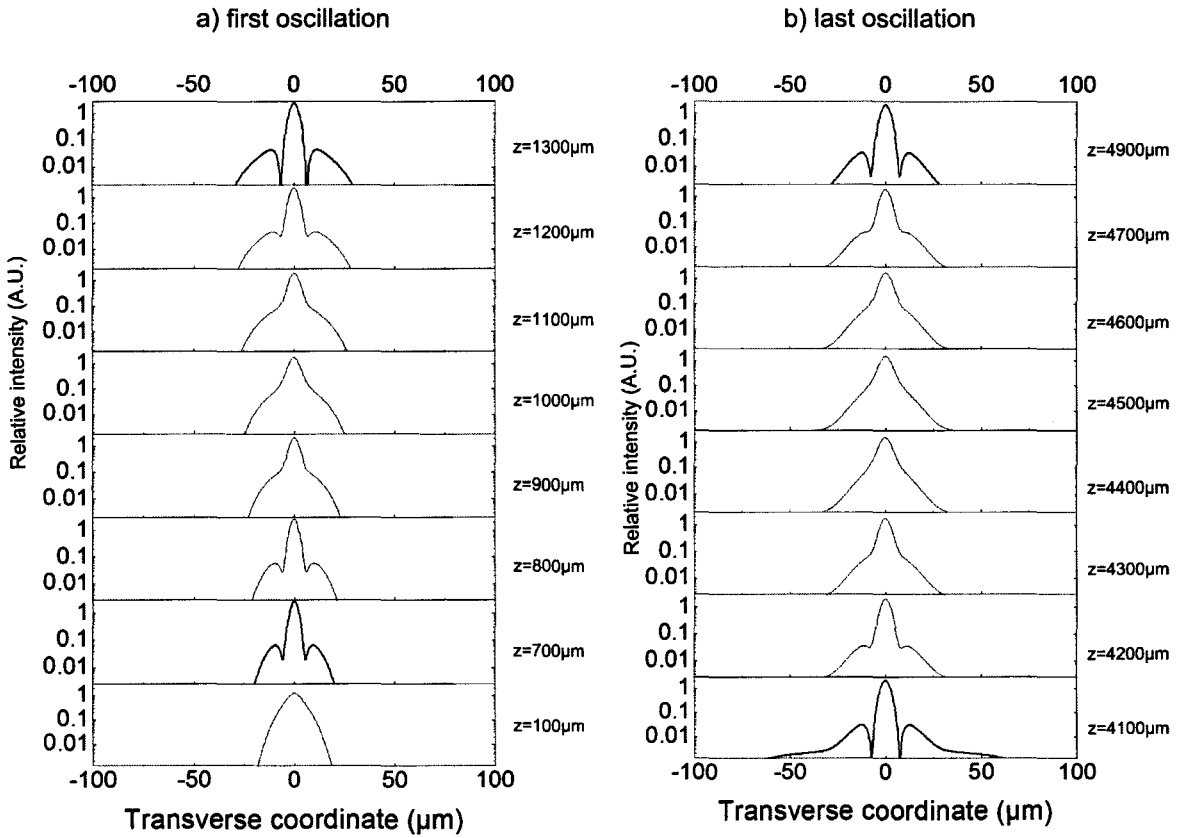


Figure 6.8: Simulated intensity profiles for the a) first and b) last oscillations along the propagation axis z for step 70 of a propagating Gaussian beam with $U_0 = 500$. In bold are indicated the profiles for the intensity maxima in the oscillation. The relative intensity is in logarithmic scale for clarity.

simulations, information at the output face of the organosiloxane is required. Figure 6.9 shows the simulated intensity profiles over time (steps) for $U_0=50$ at $z=6.00\text{mm}$.

A greater simulated intensity was chosen to make the comparison as it presented greater modal composition (see following Section). The simulated intensity variations over time (top plot in Figure 6.9) correlated well with our experimental findings at low intensities where oscillations in intensity occurred with an increase in the intensity maxima as time progressed. This was shown in the temporal plot of the evolution of peak intensity resulting from experiments at an intensity of $3.2 \times 10^{-3} \text{ W/cm}^2$ in Figure 6.10a.

In our experiments, we observed oscillations in intensity and beam width over time when monitoring at the output of the medium. These oscillations were correlated with the observation of the fundamental and higher order modes in the following way: during the intensity peaks of the oscillations the fundamental mode was dominant and during the valleys high-order modes were observed individually or superposed with other modes. This can be seen in the 2D and 1D intensity profiles in Figure 6.10b, having the intensity peaks in b1 and b6 and high-order modes in b2, b3 and b4 corresponding to LP_{02} , LP_{21} and LP_{11} . Intensity profiles in simulations showed a similar trend, where at peaks of the oscillation a dominant central intensity was observed whereas at valleys of the oscillations, higher order modes were present. This became clear at $U_0=50$ as shown in the individual intensity profiles in Figure 6.9. The intensity peaks were observed at step 36 and step 41, in between, there was one oscillation in intensity and in the valleys of the oscillations two different higher order modes were detected. Assuming radial symmetry, these modes corresponded to LP_{02} and either LP_{11} or LP_{21} in steps 38 and 39, respectively. The sequence of modes therefore agreed well with the experimental one for this particular intensity. During peaks, although the beam did not correspond to the fundamental mode in simulations but rather to a superposition of modes, the central peak was dominant in intensity compared to the side lobes. In experiments we saw most of the time only a circular beam but occasionally side lobes appeared as in b6 in Figure 6.10.

6.3.3 Dependence of self-trapping dynamics in intensity

When the input intensity of the beam was increased in simulations, and this required using lower values of U_0 , self-trapping occurred at faster rates compared to $U_0=500$, which is the lowest simulated intensity. For example, the translation of the first maxima and narrowing of the beam for $U_0=100$, 50 and 10 (see Figures C.1 and C.4 in Appendix C, 3.5 in Chapter 3) occurred at steps 5, 3 and 2 respectively, earlier than at $U_0=500$. This followed from the fact that higher intensities induce greater refractive index changes. The beam at $U_0=100$, 50 and 10 were able to induce in step 1 refractive index changes across all the propagation of the Gaussian beam (Figures C.2 and C.4 in Appendix C and 3.4 in Chapter 3) and not only in high intensity areas as it occurred at $U_0=500$. A thin waveguide was induced at faster

Profiles at $z=6.00\text{mm}$

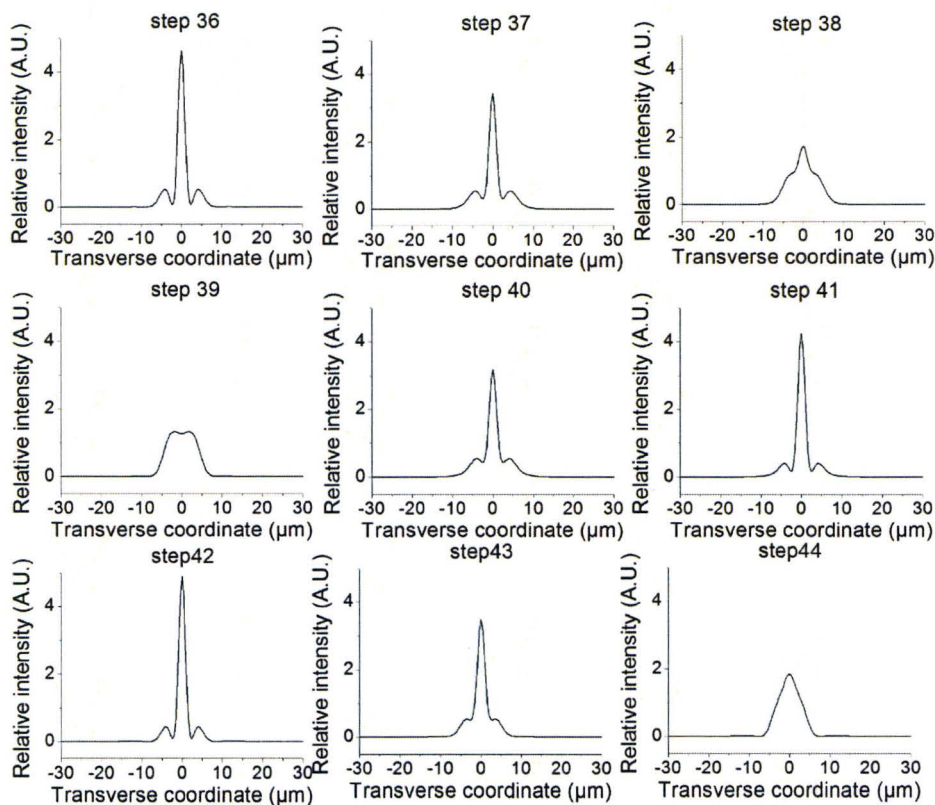
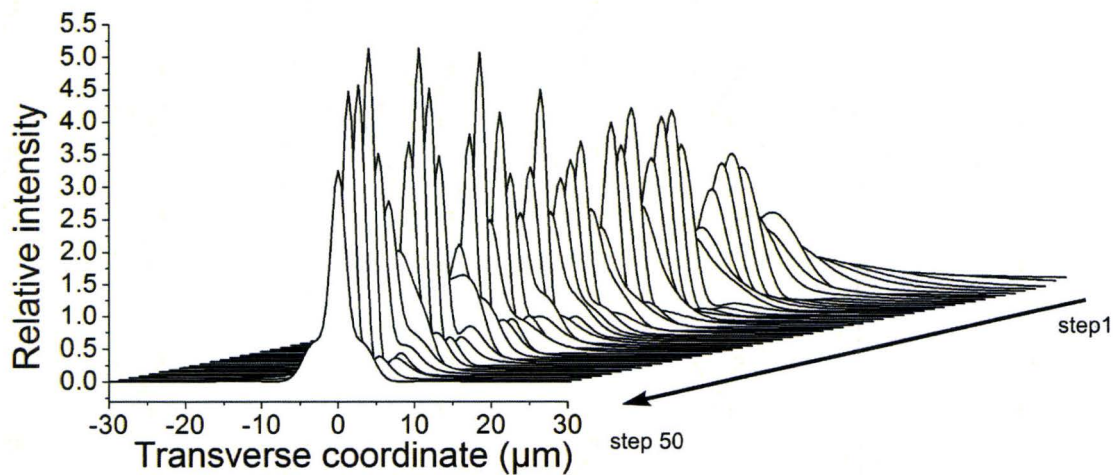
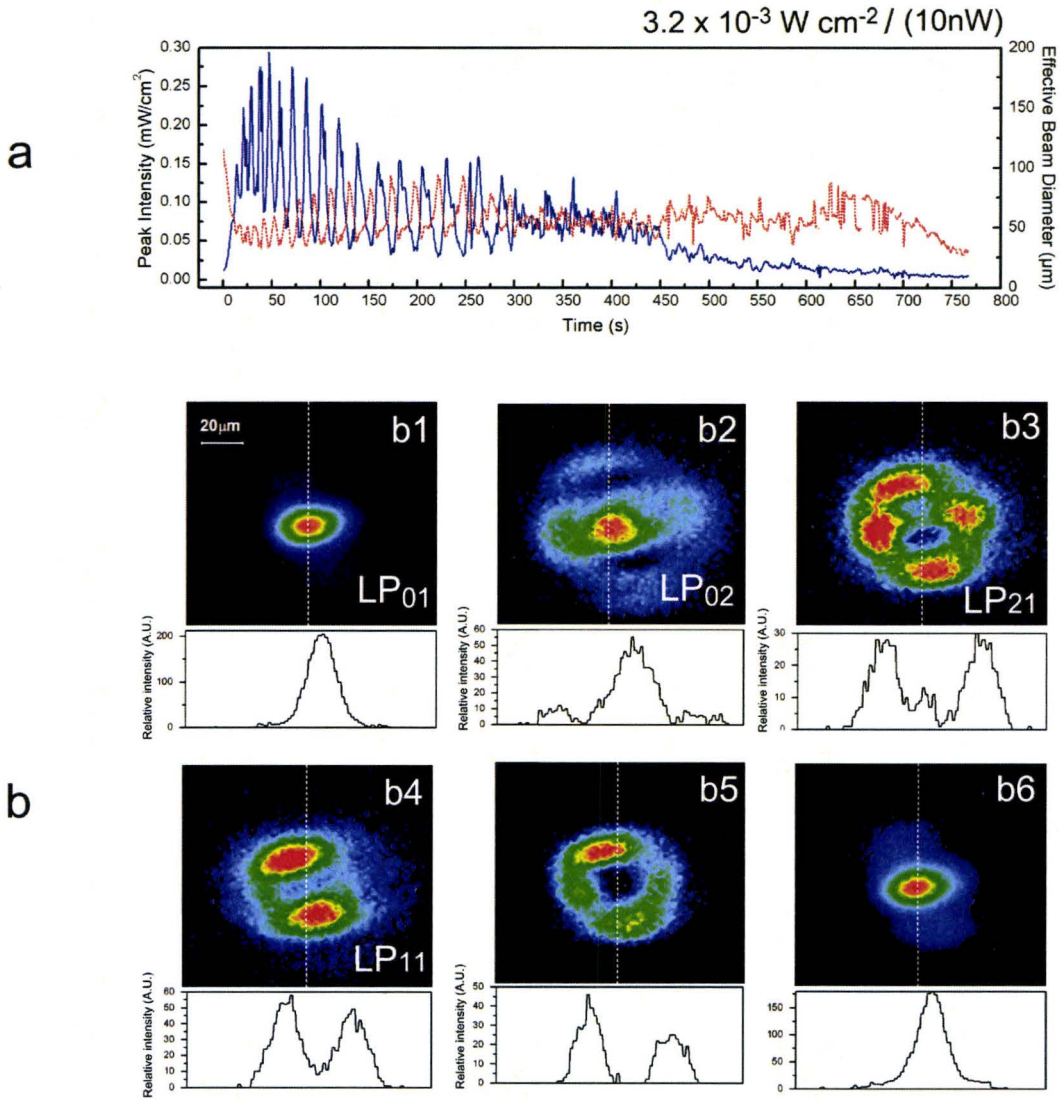


Figure 6.9: Profiles of intensity for $U_0 = 50$ in the transversal direction x at the output of the medium ($z = 6.00\text{ mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step in the computation represents a unit of time.



rates at $U_0=100$ and 50 compared to $U_0=500$. We also observed that after steps 20 and 6 for the higher intensities $U_0=50$ and 10 respectively, the waveguide experienced broadening after self-trapping. This broadening in simulations, correlates well with the broadening of the beam width observed during our experiments.

When comparing self-trapping parameters such as intensity increase, self-trapped width and self-focusing time at different U_0 in our simulations, we obtained very good agreement with trends observed in our experimental study for the low intensity regime (see Section 3.3.2). As intensity is increased in the low regime, self-trapping occurred at a faster rate and the beam experienced a faster self-focusing time. This can be observed by the maximum relative intensity occurring in our simulations at steps 68, 48, 47 and 13 for $U_0=500$, 100, 50 and 10, respectively, where the decreasing U_0 translated into increasing intensities. Moreover, an optimum intensity was found at which the self-trapping is more efficient. This was indicated by a maximum intensity transmittance and enhancement at the output face of the medium along with a minimum self-trapping beam width. This occurred in our simulations for $U_0=50$, where there was a 33-fold increase in intensity at step 47 compared to step 1 (diffracted beam) and a self-trapped beam width at step 47 of $3.18 \mu\text{m}$ as shown in Table 6.1. When comparing with the other lower and higher intensities (or U_0), the increase in intensity is smaller and the beam width is bigger. This trend was consistent with an optimum self-trapping intensity found during our experimental studies.

Table 6.1: Simulation results for low intensity regime

U_0	Intensity increase	Self-trapped beam width (μm)	Step
500	14-fold	9.07	68
100	28.3-fold	3.77	48
50	33-fold	3.18	47
10	25.3-fold	4.85	13

The intensity along z was not uniform during self-trapping for all the simulated values of U_0 . Multiple intensity maxima were present and as the intensity was increased (or lower U_0) the appearance of maxima occurred at a faster rate. Over time, more intensity maxima were observed for $U_0=100$, 50 and 10 (see Figures C.1 and C.3 in Appendix C and Figure 3.5 in Chapter 3). This resulted from the increasing polymerization rates with intensity. With increasing intensity, the modal evolution of one cycle changed drastically along z for a given step. For example at $U_0=50$ in Figure C.5c, the intensity profiles in the oscillation from $z=100 \mu\text{m}$ to $z=400 \mu\text{m}$ were less broad and possessed less side bands compared to the ones shown at an oscillation between $z=1400 \mu\text{m}$ to $z=1800 \mu\text{m}$. Over time, oscillations in intensity progressed in a similar fashion for $U_0=500$, 100 and 50, appearing more periodic along z . Oscillations in the beam for the two highest intensities became more pronounced. In

the case of $U_0=50$, this happened after step 20 (Figure 3.5 in Chapter 3) and for $U_0=10$ this was visible from step 6 (Figure C.3 in Appendix C). The strength of the oscillations was correlated with the modal profile observed along z . As the intensity was increased (lower U_0) a richer modal composition appeared during oscillations as observed for $U_0=100$, 50 and 10 in Figures C.5b, c and d in Appendix C, respectively.

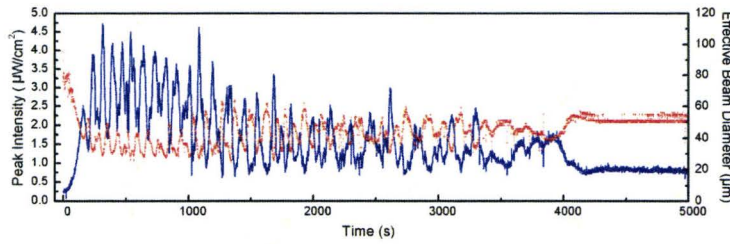
When comparing the temporal evolution of intensity at the output of the medium $z=6.00$ mm between simulations and experiments, we found a good agreement in the overall behaviour of intensity oscillations. At low intensities, the oscillations increased progressively and with certain periodicity for both experiments and simulations (Figure 6.11 a1 and b1). At higher intensities, oscillations were more irregular and had the greatest intensity peak at early times compared to lower intensities for both experiments and simulations (Figure 6.11a2 and b2). By examining the individual intensity profiles at the output of the medium over time, we saw a trend where more variation in intensity existed at higher intensities, which translated into the existence of more high-order modes compared to low intensities. This was clearly observed when comparing the profiles in Figures 6.12, 6.13, 6.9 and 6.14, corresponding to the output intensity profiles at $U_0=500$, 100, 50 and 10, respectively. A similar trend was observed during experiments in the low intensity regime, where more high-order modes occurred at higher intensities. Figure 6.15 shows selected experimental 2D intensity profiles at the highest intensity 1.6×10^{-2} W/cm² within the low intensity regime. High-order modes are shown corresponding to LP_{11} , LP_{21} , LP_{02} and superpositions of modes such as LP_{03} , LP_{02} and LP_{11} . When comparing these profiles with the simulated profiles of $U_0=10$ in Figure 6.14, assuming circular symmetry, we were able to identify LP_{11} or LP_{21} at steps 6 and 10, LP_{02} at steps 7 and 8 and a similar superposition of modes LP_{03} , LP_{02} and LP_{11} , at step 17.

The existence of high-order modes at higher intensities can be explained through the intensity dependence of the rate of polymerization and hence the rate of refractive index changes. At low intensities, a slow polymerization rate occurs compared to high intensities and therefore a smaller refractive index change is induced. At high intensities saturation is reached faster, therefore the induced waveguide broadens and more modes can be supported. The broadening is clearly observed in the refractive index profiles for $U_0=10$ in Figure C.4 in Appendix C.

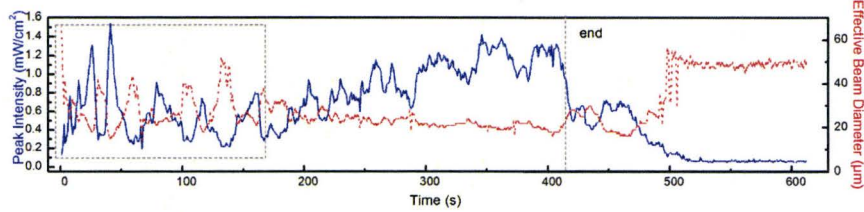
Simulations in the low intensity regime were consistent with the trends obtained in our experiments. An optimum input intensity was found during simulations where self-trapping is more efficient. Oscillations of the beam were observed along the propagation direction and at the output of the medium overtime during self-trapping. The correlation between the oscillations and the observation of high-order modes gave further insight into the evolution of modes with intensity.

a) Experimental results

a1) $3.2 \times 10^{-5} \text{ W cm}^{-2}$ / (0.1nW)

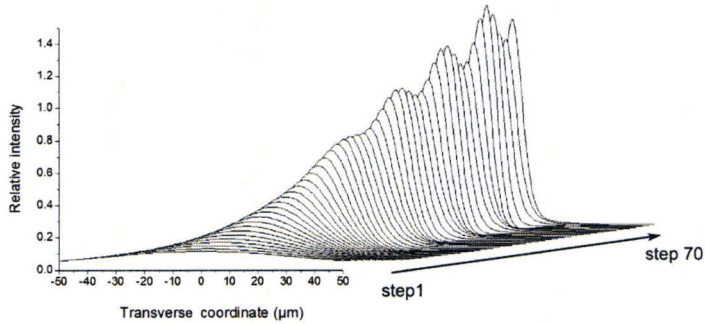


a2) $1.6 \times 10^{-2} \text{ W cm}^{-2}$ / (50nW)



b) Simulated results

b1) $U_0=500$



b2) $U_0=10$

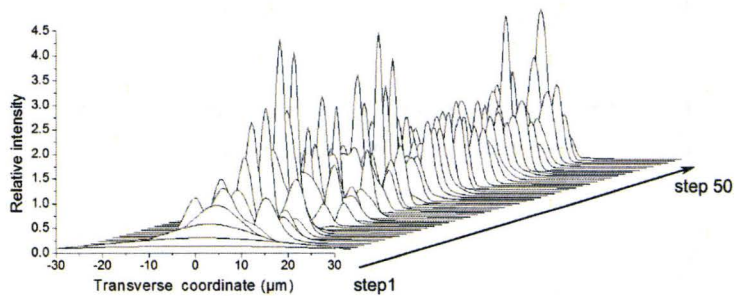


Figure 6.11: Comparison of intensity oscillations occurring overtime at the output of the medium ($z = 6.00 \text{ mm}$) between a) experimental and b) simulated results corresponding to the lowest and highest intensity within the low intensity regime. In simulations, each step represents a relative unit of time.

Profiles at $z=6.00\text{mm}$ for $U_0=500$

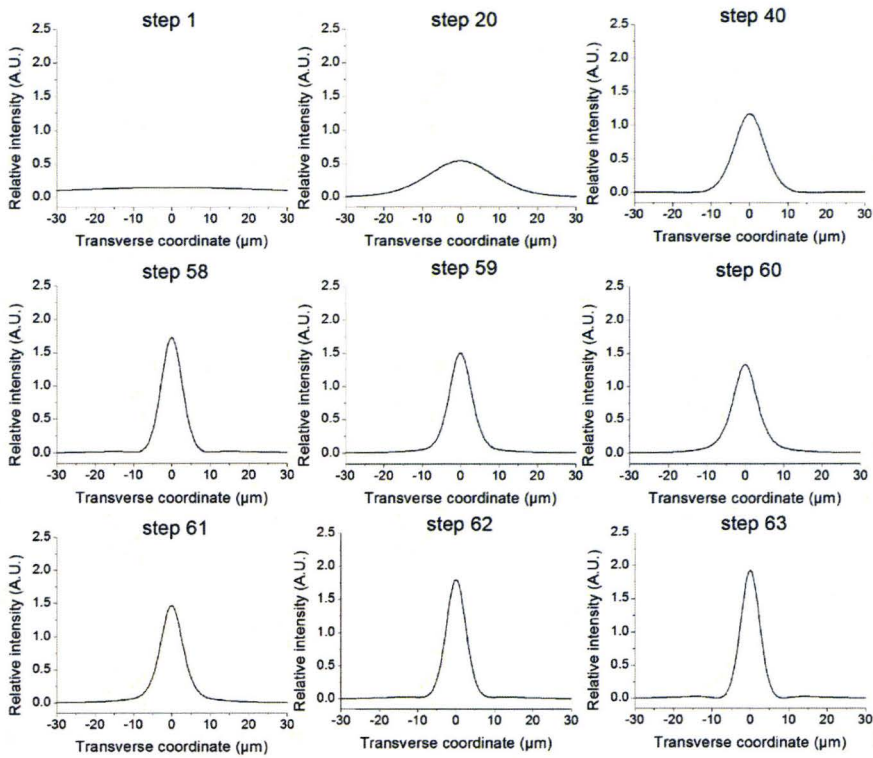


Figure 6.12: Profiles of intensity for $U_0 = 500$ in the transversal direction x at the output of the medium ($z = 6.00\text{ mm}$) for 70 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit of time.

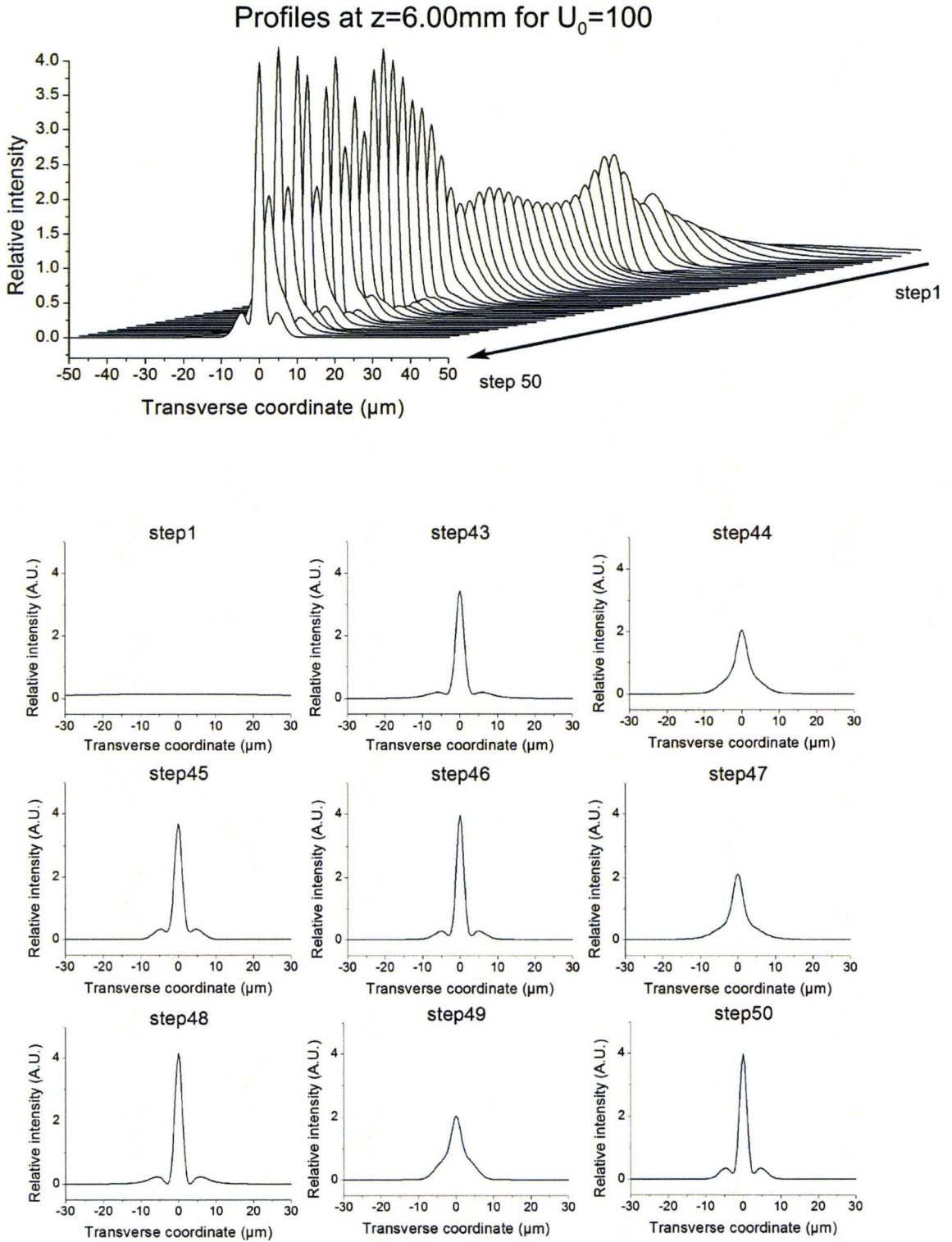


Figure 6.13: Profiles of intensity for $U_0 = 100$ in the transversal direction x at the output of the medium ($z = 6.00 \text{ mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit of time.

Profiles at $z=6.00\text{mm}$ for $U_0=10$

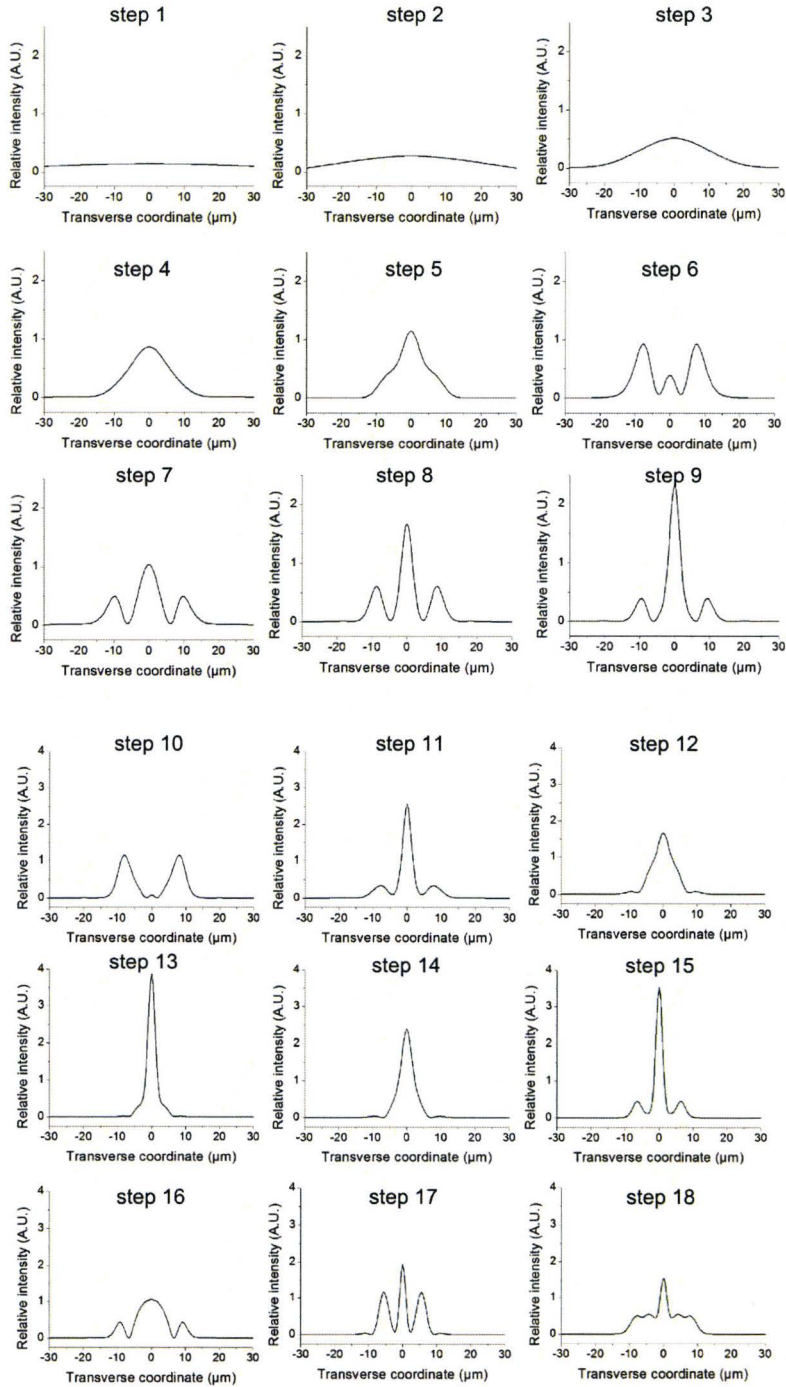


Figure 6.14: Profiles of intensity for $U_0 = 10$ in the transversal direction x at the output of the medium ($z = 6.00 \text{ mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit of time.

Experimental profiles

$1.6 \times 10^{-2} \text{ W/cm}^2$

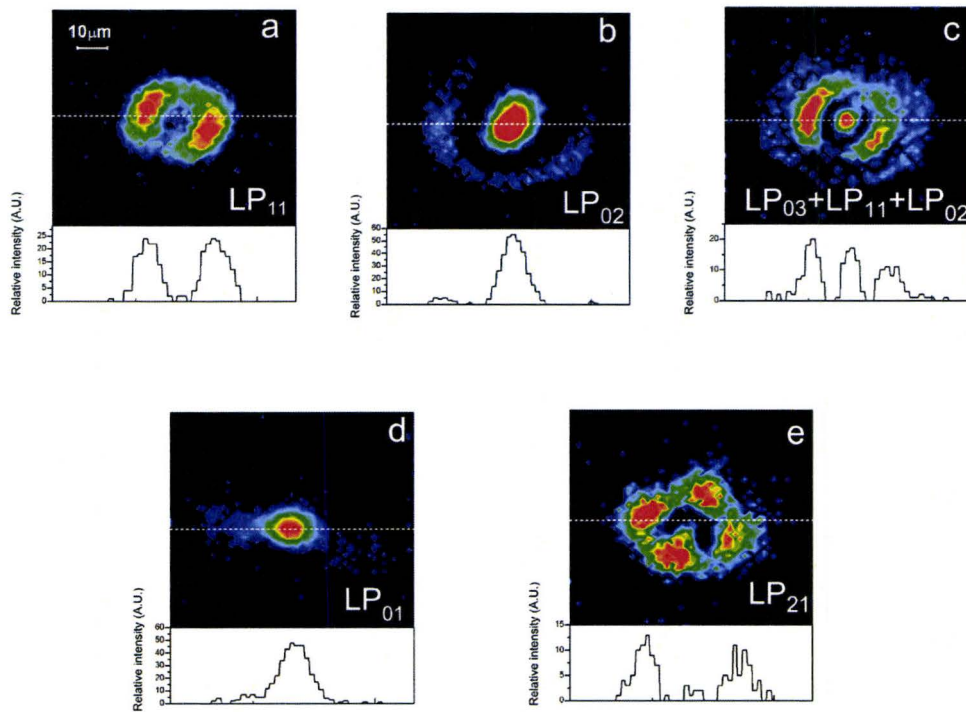


Figure 6.15: 2D experimental profiles of intensity at $1.6 \times 10^{-2} \text{ W/cm}^2$ for selected modes at (a) 39 s (b) 51 s (c) 61 s (d) 120 s and (e) 138 s with their corresponding 1D profile.

6.4 Mid-intensity regime: diffraction rings

In the mid-intensity regime, simulations showed the emergence of diffraction rings. We found that from $U_0=5$ to 0.5, diffraction rings appeared with oscillations similar to diffraction rings with high-order modes observed experimentally. At $U_0=0.1$ to 0.05 the rings dynamics are similar to dark rings. Introduction of an intensity threshold resulted in simulated dynamics of diffraction rings that are consistent with the experimentally observed dark rings.

6.4.1 Diffraction rings

At the simulated intensity of $U_0=5$, we observed the emergence of multiple lobes which can be interpreted (with radial symmetry) as rings. These were accompanied with oscillations as seen in Figure 6.16. In Chapter 4 we detailed the different ring types observed at the mid-intensity regime, we found intensity profiles which present diffraction rings superposed with high-order modes occurring with oscillations of the beam. Simulations at $U_0=5$ show a similar behaviour to this type of rings.

Figure 6.16 shows the intensity profiles at the output face of the medium for various steps (or time) for $U_0=5$. We observed between step 3 and step 9, an oscillation in intensity composed of intensity profiles that with radial symmetry, are very similar to the experimental results for rings with high-order modes shown between 13 to 20 seconds in Figure 6.17. There, we observed a sequence of a bright beam surrounded by one ring at 13 s and a single ring with a small central peak at 17 s. At later steps in the simulations, (Figure 6.18) such as step 10 and 13, more rings appeared and oscillations continued varying. A similar behaviour was observed in the experimental results in Figure 6.17 between 24 to 40 s where more rings appeared as oscillations occurred. The intensity peaks of the oscillations in the simulations, appeared with different number of surrounding rings as seen in step 11 and 17 in Figure 6.18. During experiments, we observed a similar behaviour mainly at later stages as seen at around 49, 84 and 162 s in Figure 6.17 where the intensity peaks have multiple rings surrounding it.

Figure 6.19 shows the intensity and refractive index maps for simulations at $U_0=5$ for various steps. The intensity map shows an intricate evolution along the propagating direction z over time. At step 5 we can observe that the beam already presents very different profiles at various z cross sections, as time evolves (steps), the beam narrows slightly but still preserving the intricate intensity profiles across z . The refractive index maps show that at early stages (step 2 and 3), the refractive index profile at the entrance of the medium remains Gaussian, however as simulations evolve over time, the profile saturates at the entrance of the medium creating a top-hat index profile. The overall structure takes a conical form which correlates well with structure induced during experiments.

Profiles at $z=6.00\text{mm}$ for $U_0=5$

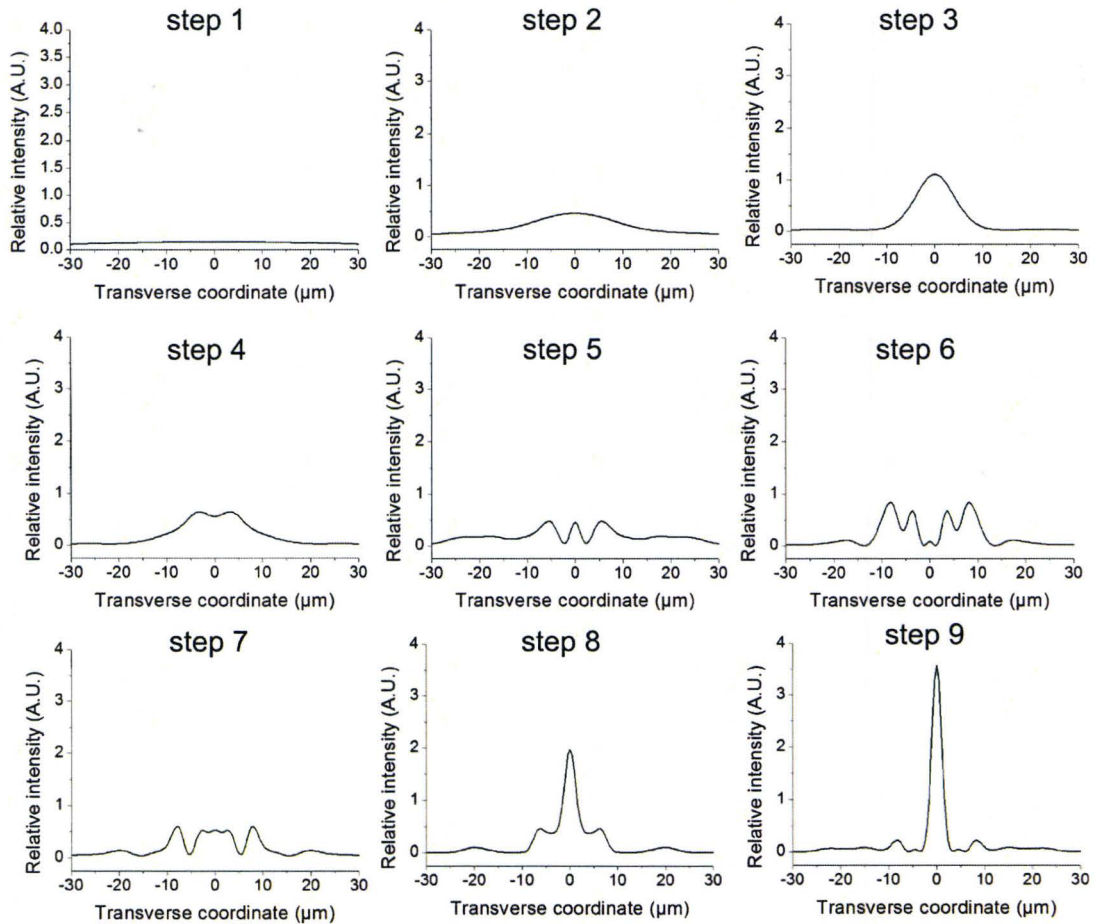
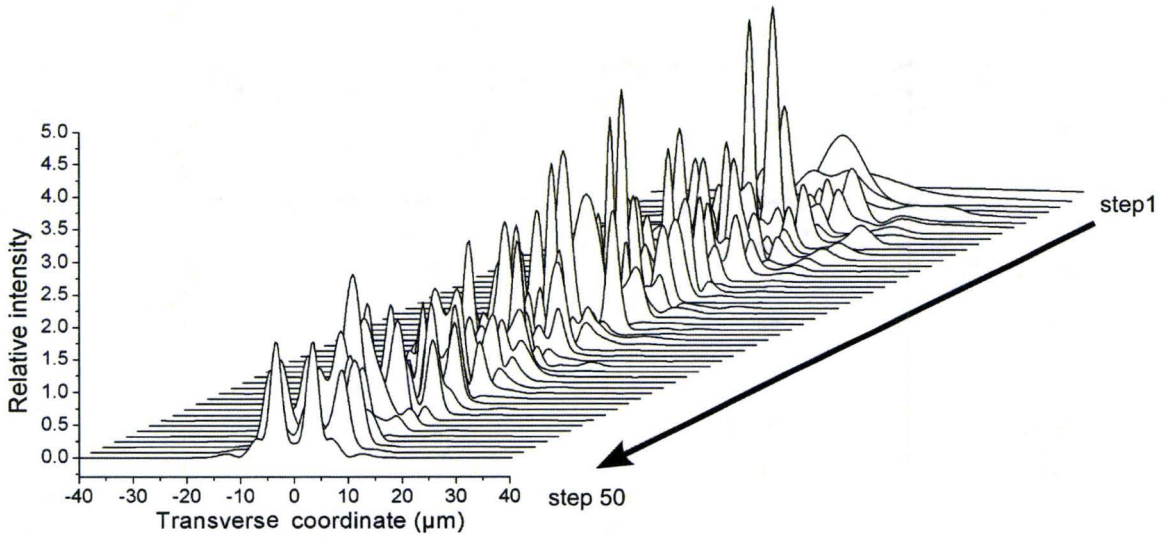


Figure 6.16: Profiles of intensity in the transversal direction x at the output of the medium ($z = 6.00 \text{ mm}$) for 50 computation steps obtained in Beamprop simulations for $U_0 = 5$.

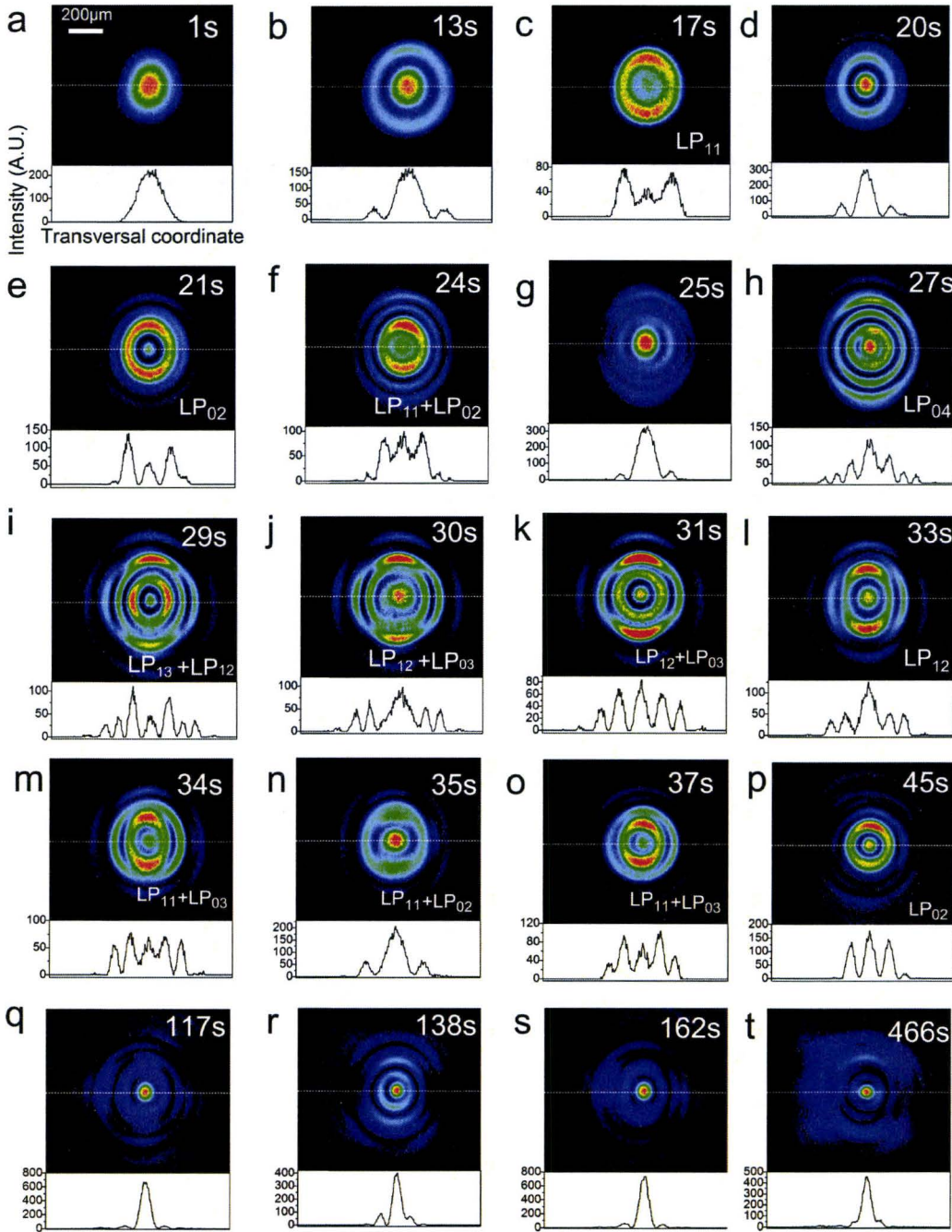


Figure 6.17: Experimental results of 2D and 1D spatial intensity profiles at 1.6 W/cm^2 showing the evolution of diffraction rings with high-order modes.

Profiles at $z=6.00\text{mm}$ for $U_0=5$

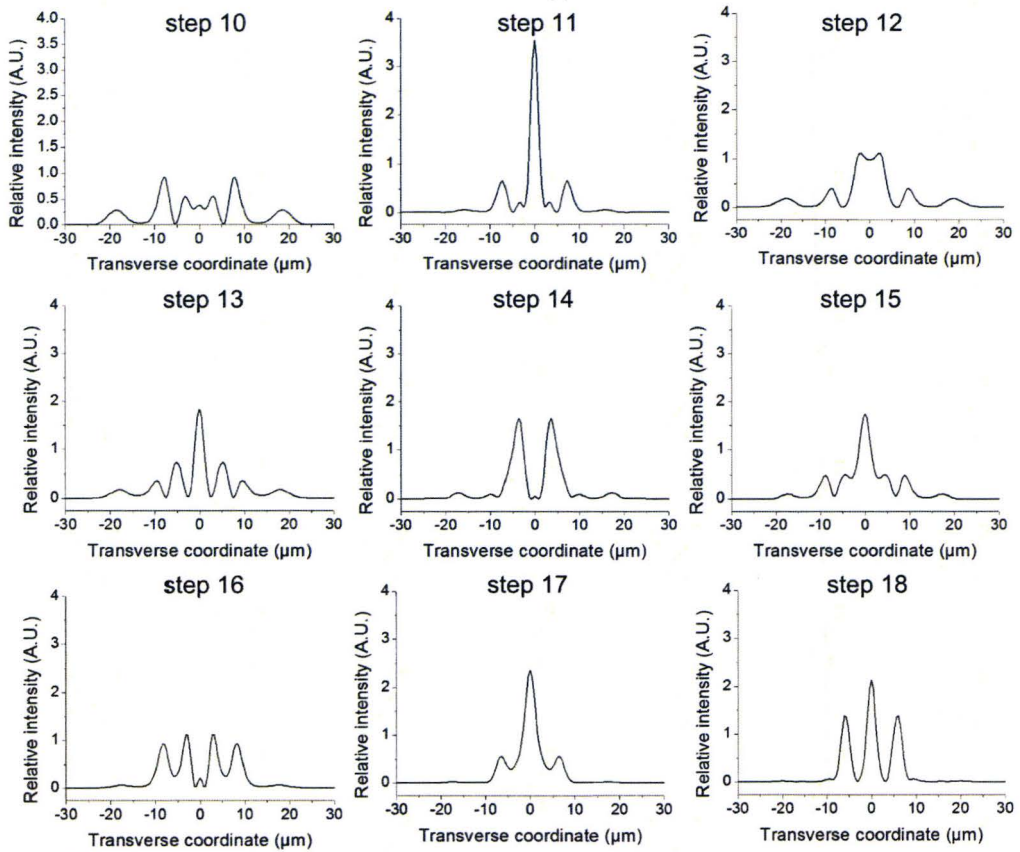


Figure 6.18: Profiles of intensity in the transversal direction x at the output of the medium ($z=6.00\text{ mm}$) for 50 computation steps obtained in Beamprop simulations for $U_0 = 5$.

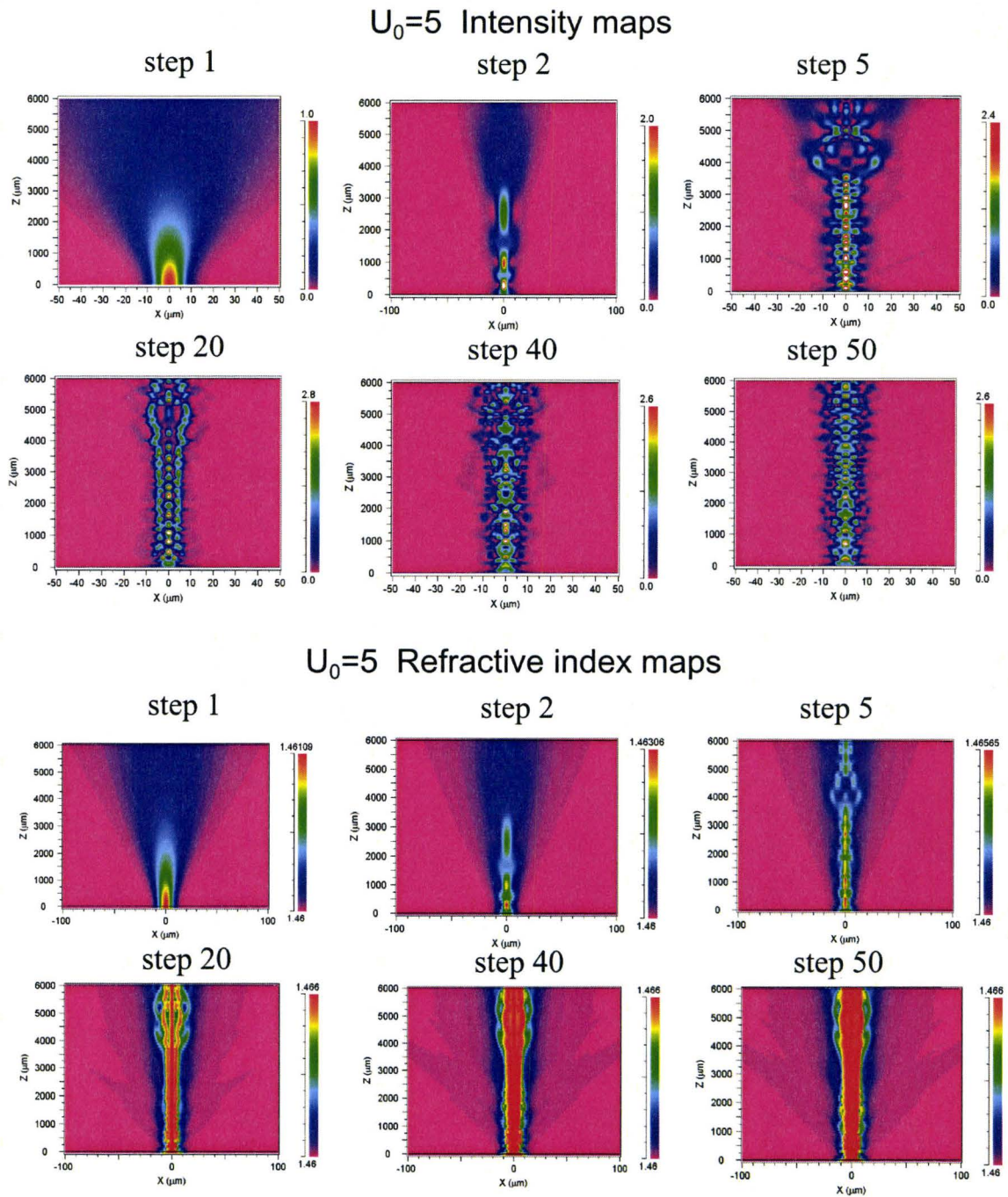


Figure 6.19: 2D Simulation results in Beamprop showing the intensity profiles (top) at $U_0 = 5$ of a propagating Gaussian beam with the corresponding refractive index profiles (bottom). The propagation coordinate is indicated with z and the transverse coordinate is x .

The emergence of rings and oscillations have been observed in theoretical studies of saturable self-focusing media [132]. The appearance of rings with oscillations were described to correspond to a different family of light patterns compared to the one with linear polarization in x or y to high-order modes [114]. The fact of having a different family of light patterns could originate from increasing intensity in the mid-intensity regime compared to the low intensity regime creating different propagating dynamics. This could explain the observation of rings with high-order modes in experiments and simulations.

When intensity is increased in simulations, we observed at $U_0=0.1$ the emergence of multiple rings in the first steps followed by a narrowing of the beam at later steps as seen at the output profiles at $z = 6$ mm in Figure 6.20.

This can be seen with radial symmetry in the two, four and eight lobes developed at steps 4, 8 and 12, respectively, followed by a central peak increase at step 50. Figure 6.21 shows the intensity maps for the same U_0 where we see some diffraction effects along the pathlength in steps 20 to 50. The refractive index profile at the entrance of the medium shows from step 1 the saturation of the refractive profile with a top-hat profile. The resulting structure has a conical shape similar to the one observed in experiments. The behaviour of simulations at $U_0=0.1$ follows a similar trend compare to the dark central rings observed in experimental studies (Figure 6.22), where increasing number of rings emerge. However, in the simulations a sharp peak occurs at the beginning of the experiment in step 6 of Figure 6.20, which does not correlate with experimental observations. Also the increase in intensity towards the end of experiments is sharper than the one observed in simulations. These disagreements could be due to the approximated values of critical parameters used in simulations such as maximum refractive index change Δn_s and the physical critical exposure value U_0 for polymerization. This last parameter is used to convert intensities used in experiments to U_0 values in simulations.

6.4.2 Introducing an intensity threshold

We obtained a better agreement with experimental observations of dark diffraction rings by introducing an intensity threshold. In this case we simulated for $U_0=0.05$ the diffraction rings in 2D by adding a threshold intensity which discarded the intensities below a relative value, that was chosen to be 0.5 (note that the maximum peak intensity of input beam is 1). A sequence of intensity maps resulting from simulations are shown for various computational steps in Figure 6.23. The emergence of multiple bands of intensity is shown at step 2 followed by self-trapping of the beam at around step 18. Resulting intensity profiles in Figure 6.24 at the output face ($z=6$ mm) confirm the increasing number of bands as seen from step 2 to step 10, these graphs are similar to the cross sections observed for central dark rings in Figure 6.22, therefore radial symmetry would indicate the emergence of diffraction rings. At step 13, the beam starts self-focusing, increasing the intensity peak at steps 14 and 16 until the

Profiles at $z=6.00\text{mm}$ for $U_0=0.1$

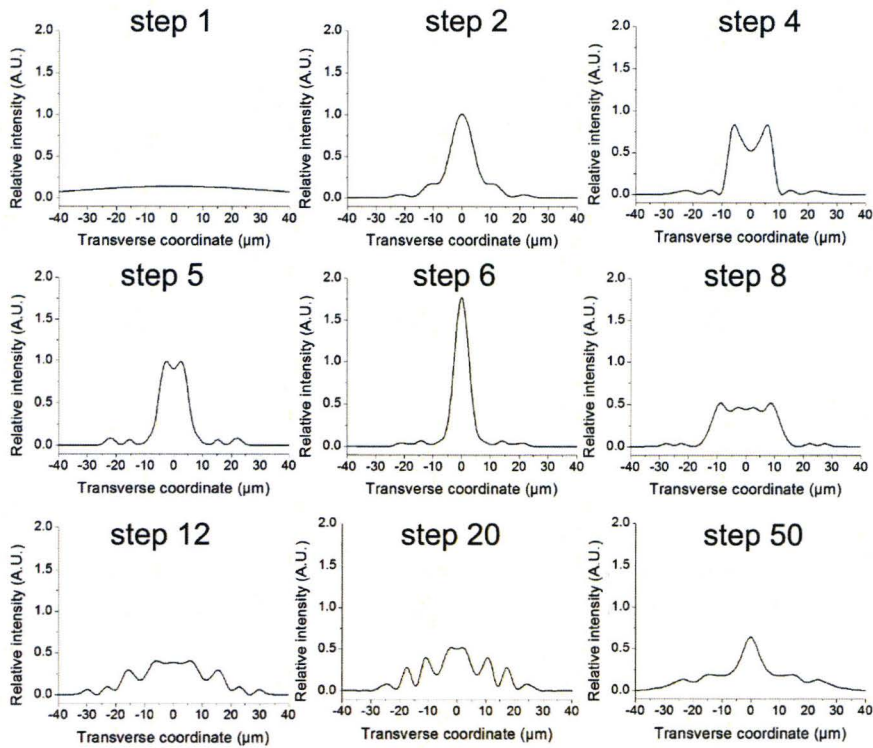
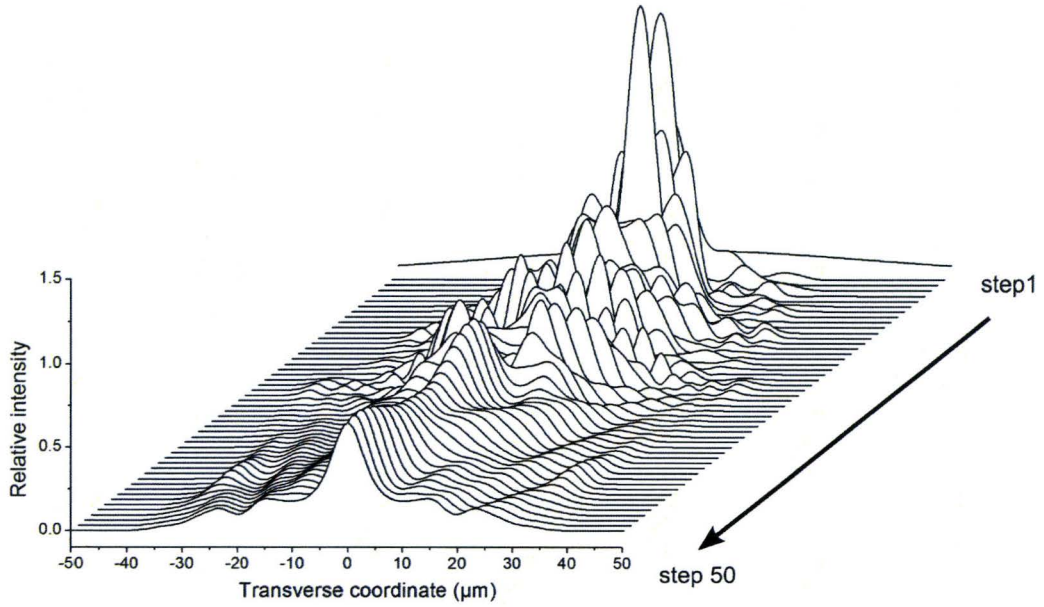
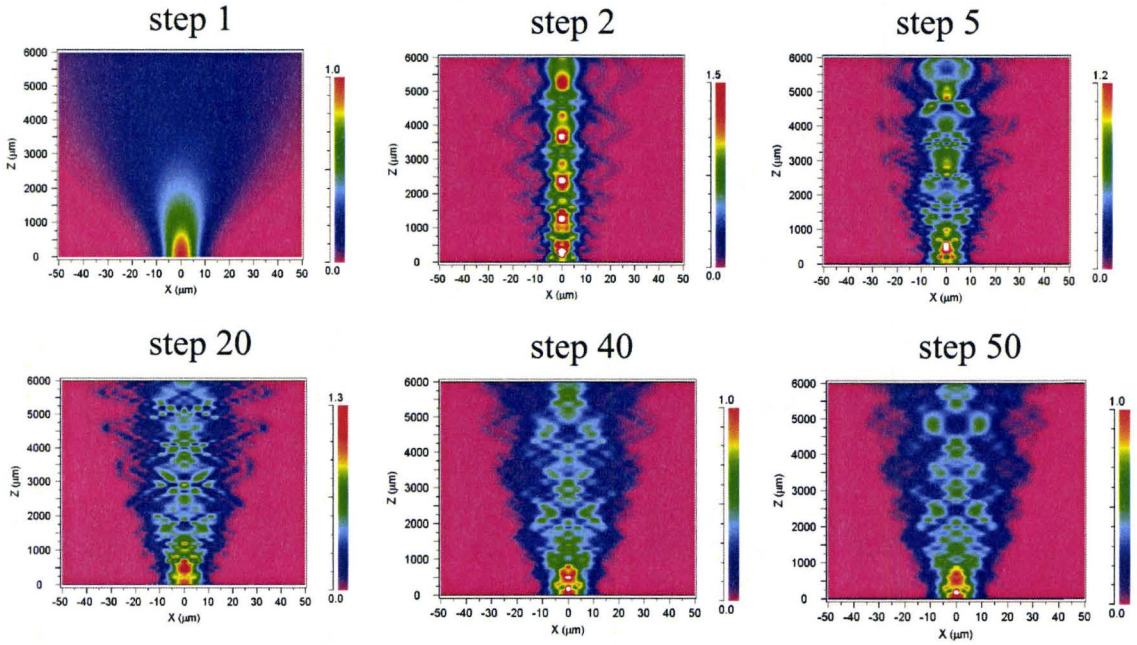


Figure 6.20: Profiles of intensity in the transversal direction x at the output of the medium ($z=6.00\text{ mm}$) for 50 computation steps obtained in Beamprop simulations for $U_0 = 0.1$.

$U_0=0.1$ Intensity maps



$U_0=0.1$ Refractive index maps

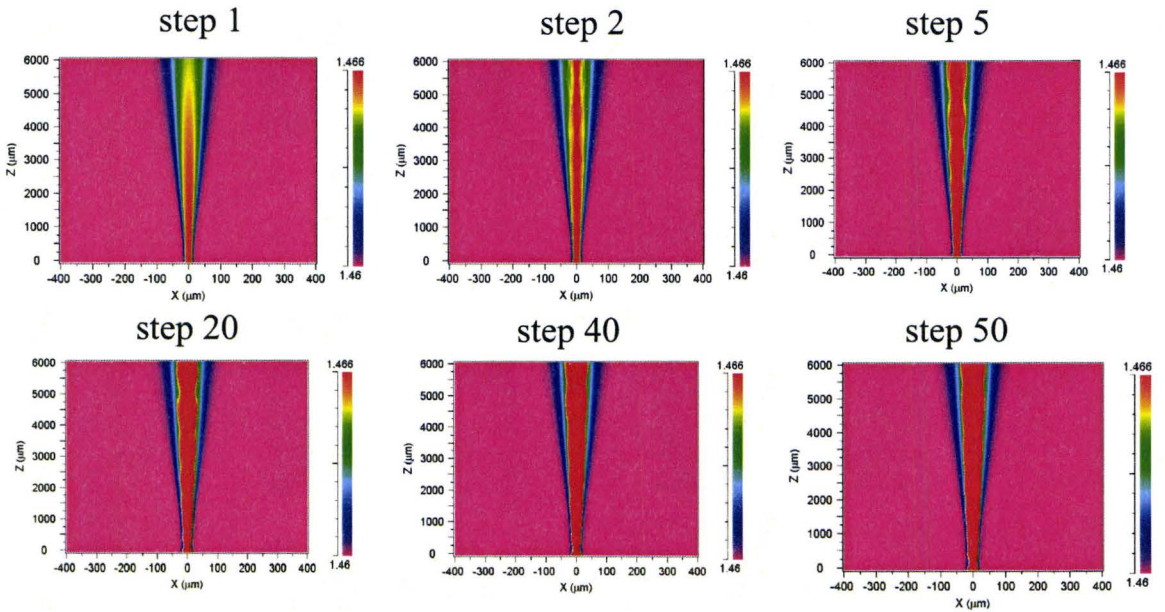


Figure 6.21: 2D Simulation results in Beamprop showing the intensity profiles (top) at $U_0 = 0.1$ of a propagating Gaussian beam with the corresponding refractive index profiles(bottom). The propagation coordinate is indicated with z and the transverse coordinate is x .

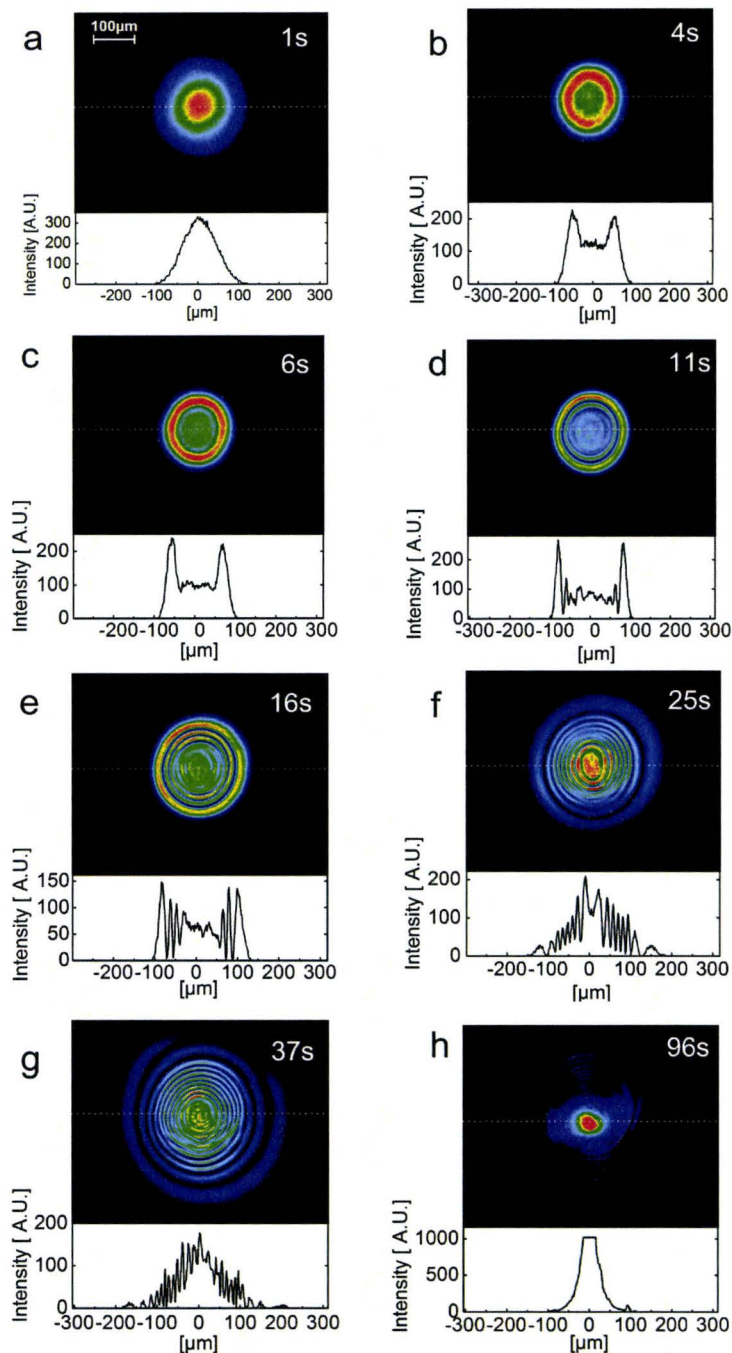


Figure 6.22: Experimental results of 2D and 1D spatial intensity profiles at 1.6 W/cm^2 showing the emergence of diffraction rings followed by self-trapping of the beam.

beam narrows at step 50. This behaviour is identical to the one observed in dark diffraction rings. However, the artificial introduction of an intensity threshold can only be justified if in experiments, the most significant refractive index changes occur at the entrance face of the medium as seen in the refractive index maps resulting from simulations in Figure 6.25. In step 1, we observe only a bullet like structure that continues growing over the following steps until reaching the other side of the material in step 16. However, experimentally the typical resulting structure has a conical shape.

The transverse refractive index profile at the entrance face saturates from the first step but it only occurs at the beginning of the sample. In the proposed mechanism in our experimental studies, a Gaussian profile is required for diffraction rings to occur. In this mechanism, calculations performed for thin media (thickness $<$ Raleigh range) only took into account that relevant refractive index changes occurred at the entrance face of the medium with linear propagation in the remaining length of the propagation medium. Simulations for $U_0=0.05$ with threshold 0.5 show that relevant refractive index changes only occurred at the entrance of the medium for early stages (step 1 to 6 of Figure 6.25). As the beam increases in intensity, the refractive index becomes more important in the remaining length of the medium as seen from the growth of the bullet-like structure from step 2 onwards (Figure 6.25). Although the refractive index profile saturates at the entrance face, the profile in the last part of the index structure seems to have a transverse gradient which could explained the emergence of diffraction rings from the tip of the structure as seen in Figure 6.23.

Previous simulations of diffraction rings for long pathlengths have been performed in a photorefractive crystal for a self-defocusing nonlinearity [80]. Diffraction rings are described as optical shock waves and simulations are performed for collisions of two of such waves. However, simulated results showed output intensity profiles only and internal dynamics are not shown due to the complexity of the simulation.

Although we observe the emergence of dark diffraction rings in simulations and rings with high-order modes at different intensities, experimentally we observe both of them at the same input intensity. A possible reason for this discrepancy is that since experimentally is difficult to precisely control the degree of polymerization, rings with high-order modes could be taking place at a different degree of polymerization compared to dark diffraction rings. Therefore, in simulations, having the same degree of polymerization and hence the same maximum refractive index change (Δn_s), the difference would be translated in the input intensity needed.

6.5 High intensity regime: single ring formation

In the high intensity regime, simulations showed the formation of a single ring for $U_0=0.005$ to 0.001, which is in agreement with our observations in experiments. At the highest intensity of $U_0=0.00001$, no significant changes occurred in the beam.

$U_0=0.05$ Intensity maps
with intensity threshold 0.5

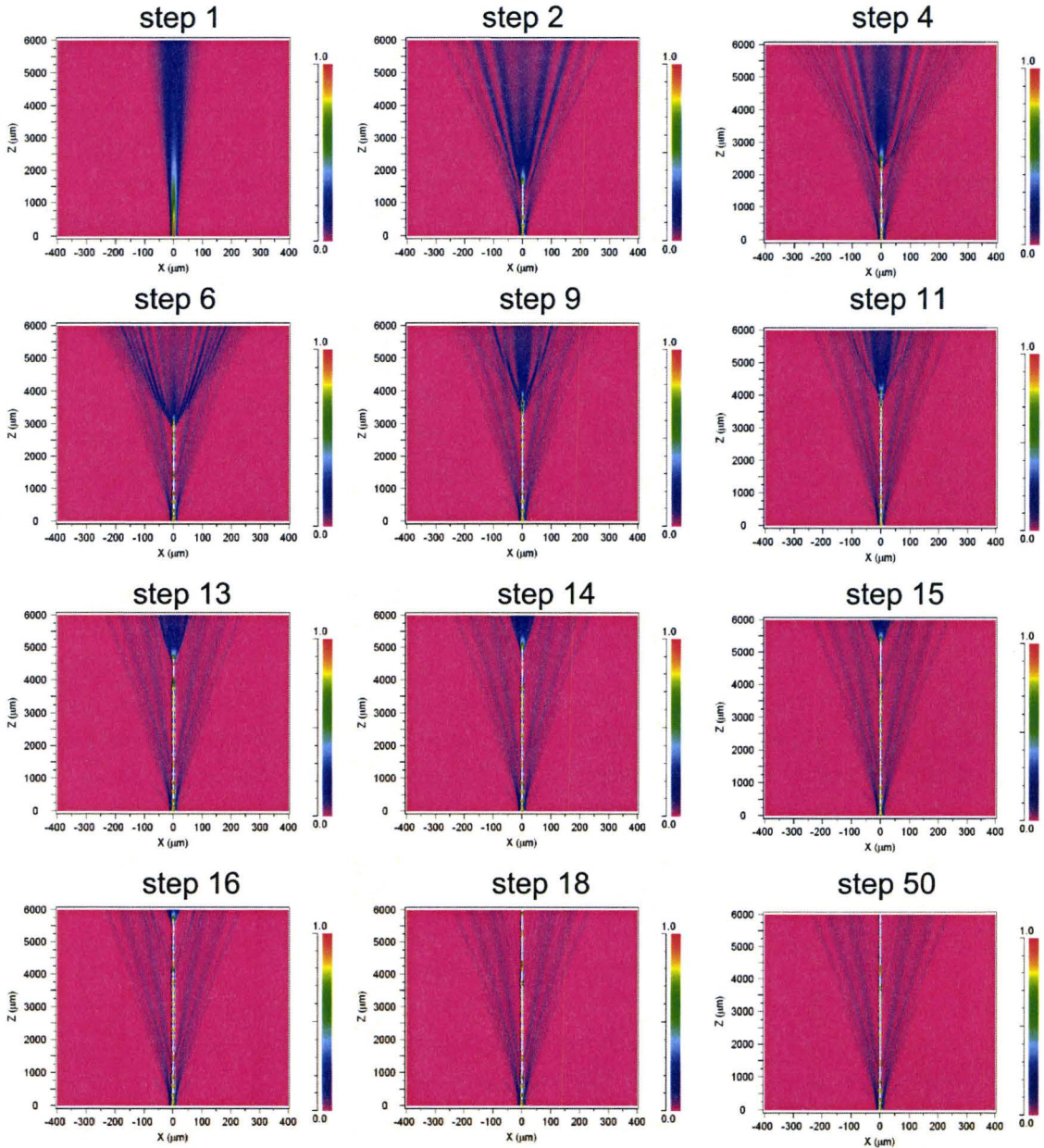


Figure 6.23: Simulations showing the emergence of diffraction rings followed by self-trapping of the beam for 50 computational steps.

Profiles at $z=6.00\text{mm}$ for $U_0=0.05$
with intensity threshold 0.5

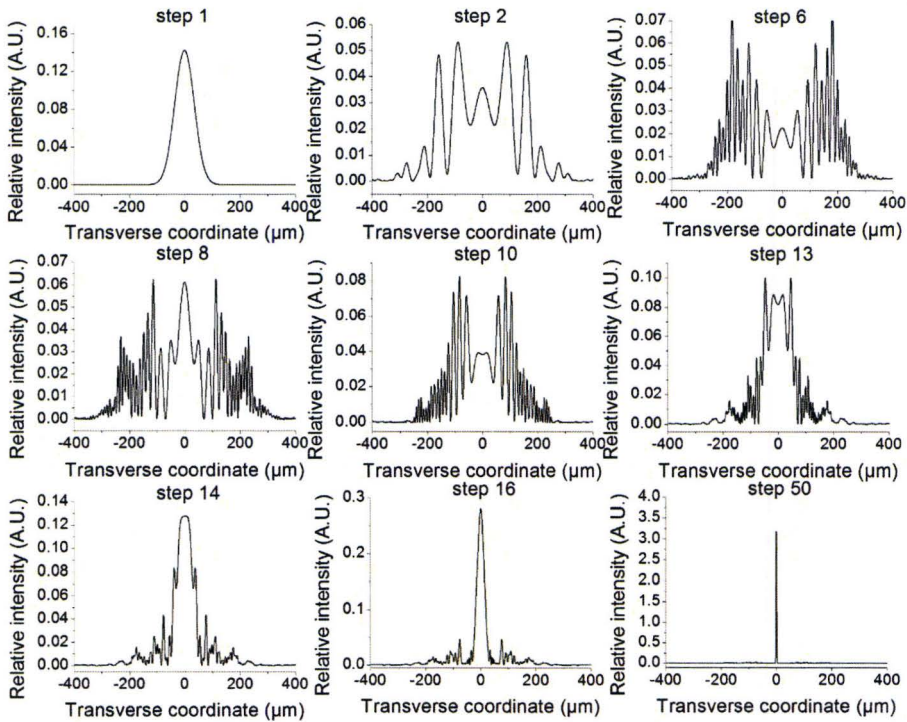
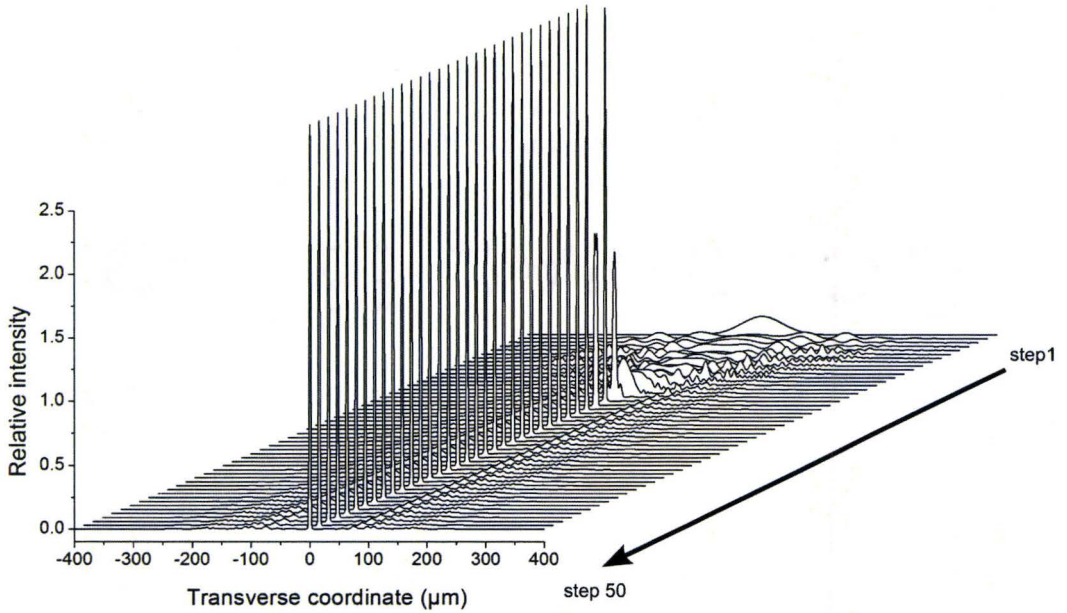


Figure 6.24: Profiles of intensity in the transversal direction x at the output of the medium ($z=6.00\text{ mm}$) for 50 computation steps obtained in Beamprop simulations for $U_0 = 0.05$. Note the different intensity scales in the individual profiles.

$U_0=0.05$ Refractive index maps
with intensity threshold 0.5

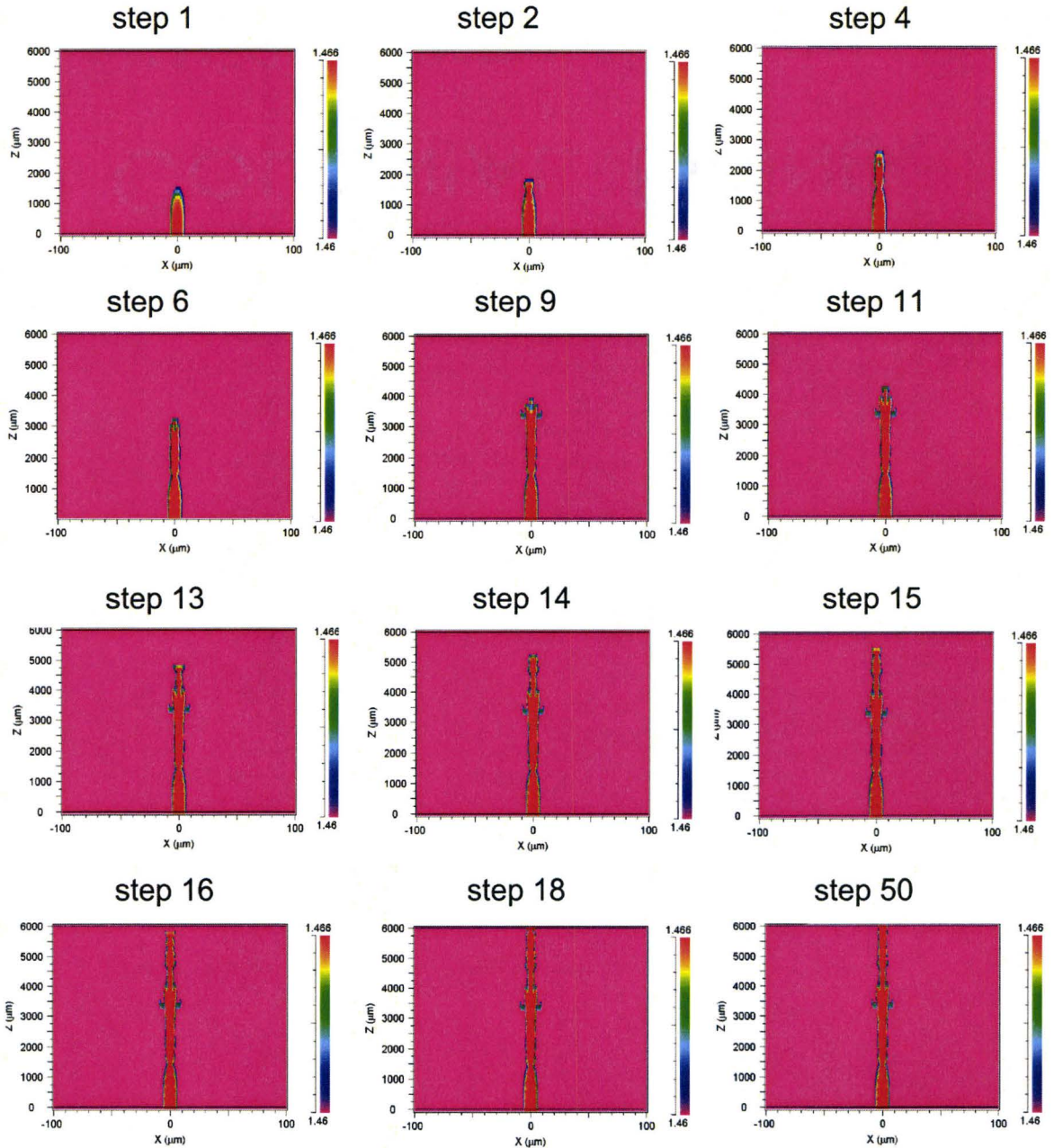


Figure 6.25: Simulations showing the refractive index maps for $U_0 = 0.05$ with threshold 0.5 for 50 computational steps.

6.5.1 Single-ring formation

We observed the single-ring formation over time at $U_0 = 0.005$ and $U_0 = 0.001$ in simulations. The results for simulations at $U_0 = 0.005$ were presented in Section 5.4. For $U_0 = 0.001$ Figure 6.28 shows at the top, the intensity profiles at $z = 6.00$ mm for 50 steps.

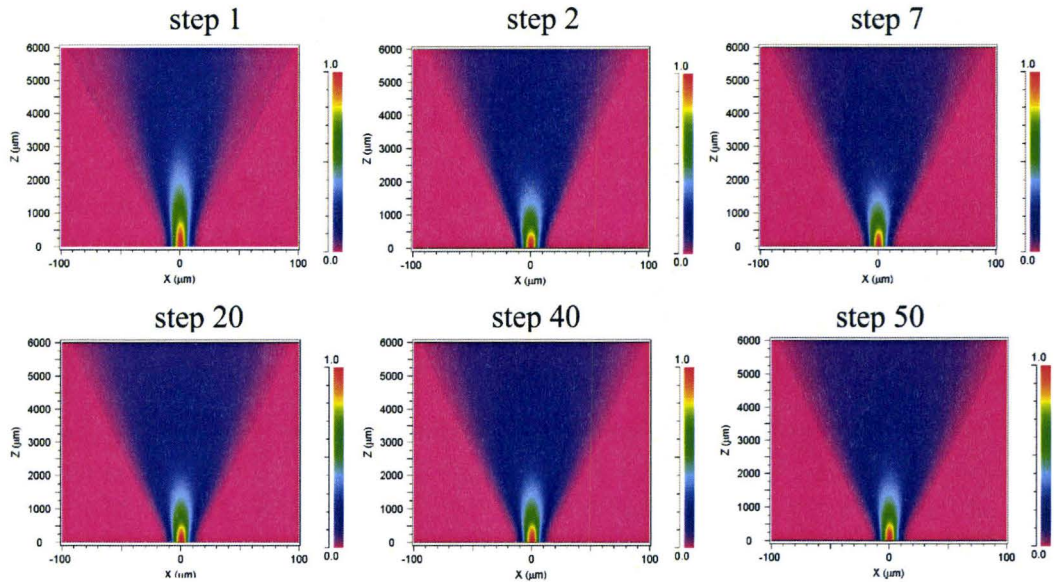
In the first steps, two and then multiple lobes are observed (step 5 to 30), taking into account radial symmetry this could translate into diffraction rings similar to the ones observed at the beginning of our experiments where single-ring formation is observed as shown in Figure 6.28b. Towards step 40 in the simulations, only two lobes are formed which again can be interpreted as the single ring formation with radial symmetry observed in our experiments in Figure 6.28f. The intensity maps along the propagation direction (z) in Figure 6.26, strengthens our interpretation, since at early steps (step 2 and 7) the beam shows diffraction of the beam along the propagation distance and at steps 40 and 50, the beam splits into two directions, which follows the propagation of flattened-Gaussian beams and Super-Gaussian beams. The refractive index maps shown in Figure 6.26 indicate that the profile at the entrance of the medium saturates in the first step, creating a flattened refractive index profile. This type of profile as shown in Section 5.3 results in single ring formation during nonlinear propagation.

6.5.2 Highest intensity

At very high intensities, simulations showed similar behaviour as the one observed in experiments. Intensity of the input beam was so high that it induced the maximum refractive index change throughout most of the width of the beam resulting in no refractive index gradient along the cross section of the beam. This prevented the formation of an initial lens at the beam waist of the Gaussian beam and it subsequently inhibited the self-focusing and self-trapping of the beam. Figure 6.29 shows the resulting intensity and refractive index profiles of 2D simulations for 50 steps. The intensity maps did not show significant changes over time for the propagating beam, except a spread in the beam. The induced refractive index profiles saturate over almost all the width of the Gaussian beam and over time the saturation extends creating a wider structure.

When monitoring the intensity beam profile at the output of the medium over many steps or time in the simulations for $U_0=0.00001$ (Figure 6.30) we observed that the beam broadened over time from step 1 to step 2. The broadened beam remained relatively unchanged over the following steps therefore no self-trapping occurred at high intensities in the simulations. This correlates well with our experimental results, since the beam was not able to overcome diffraction due to a lack of refractive index contrast in the cross section of the beam. This can clearly be observed in the bottom graphs of Figure 6.29 where the refractive index is uniform almost across all the beam.

a) $U_0=0.001$ Intensity maps



b) $U_0=0.001$ Refractive index maps

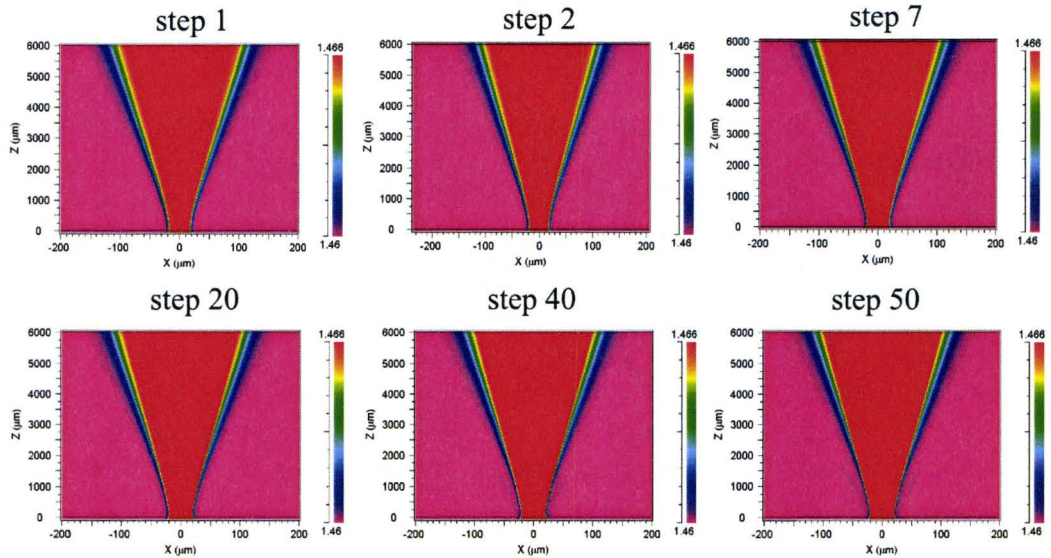


Figure 6.26: 2D Simulation results in Beamprop showing the intensity profiles (top) at $U_0 = 0.001$ of a propagating Gaussian beam with the corresponding refractive index profiles (bottom). The propagation coordinate is indicated with z and the transverse coordinate is x .

Profiles at $z=6.00\text{mm}$ for $U_0=0.001$

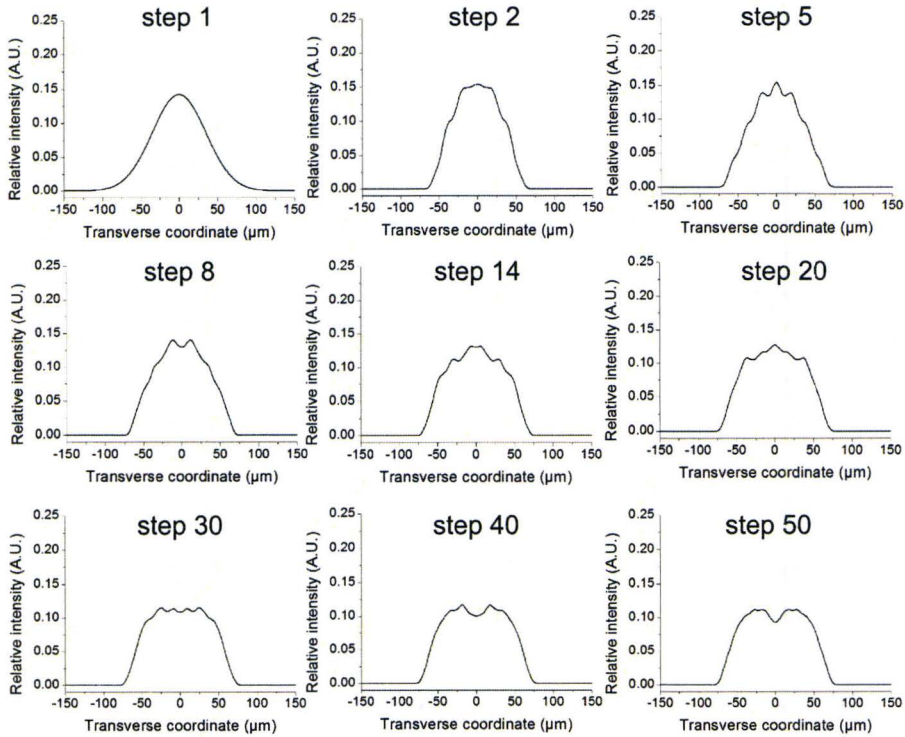
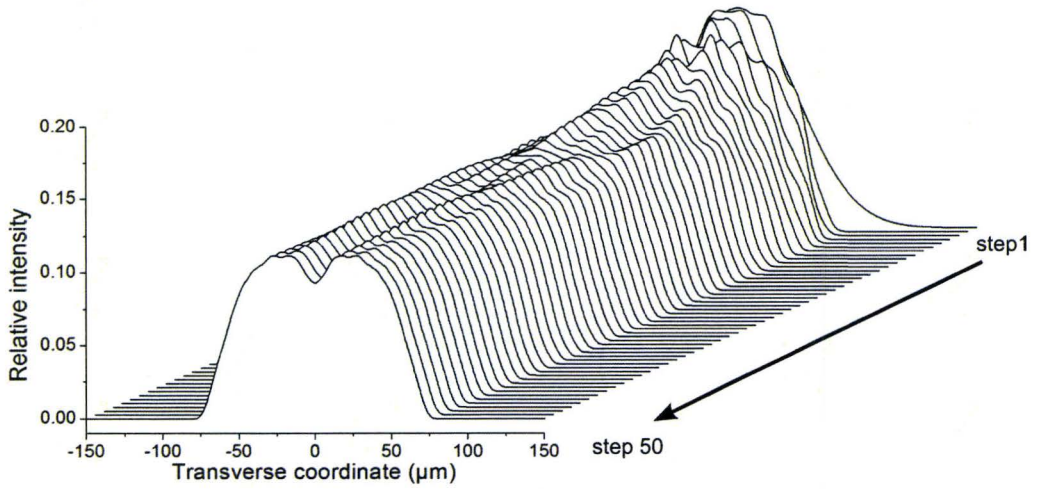


Figure 6.27: Profiles of intensity for $U_0 = 0.001$ in the transversal direction x at the output of the medium ($z=6.00\text{ mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit of time.

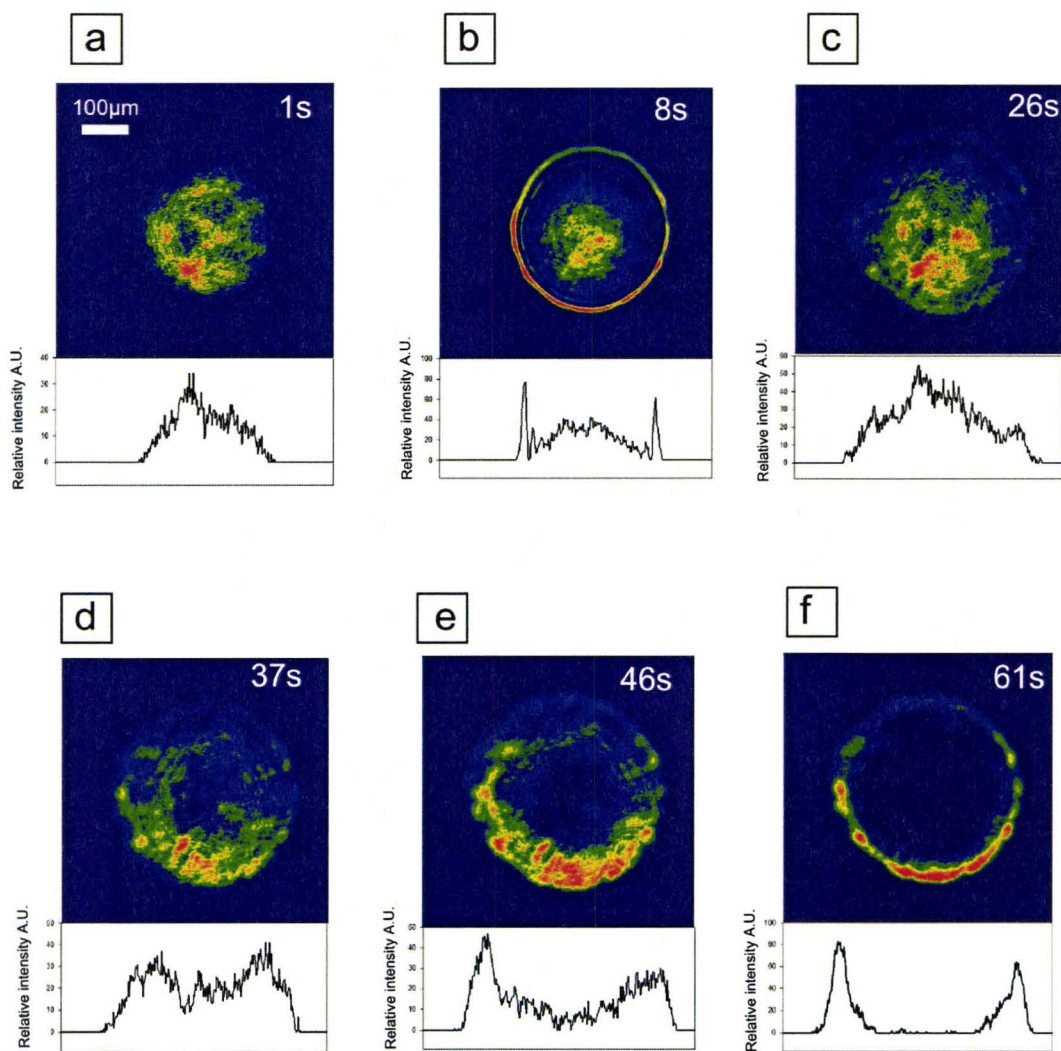


Figure 6.28: 2D and 1D experimental profiles for intensity 80 W/cm^2 showing the formation of a single ring.

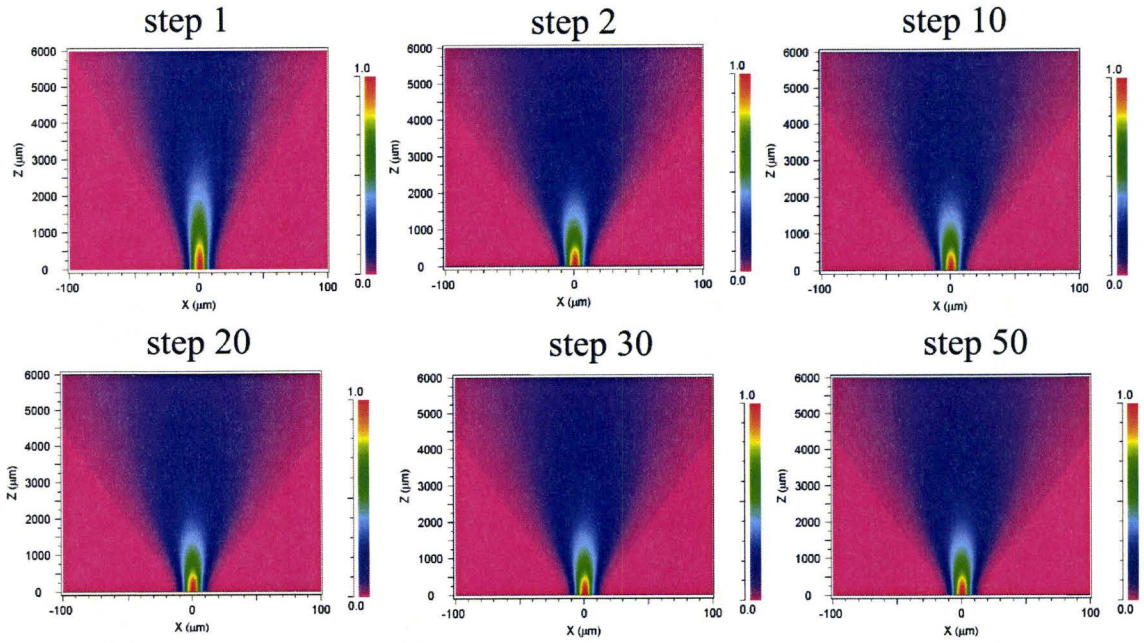
In these simulations, filamentation of the beam is not observed because we did not introduce noise in the system or the beam which is required. Further development of simulations with noise are necessary to simulate filamentation of the beam.

6.6 Conclusions

Numerical simulations of the nonlinear propagation of light in the organosiloxane were performed by combining the beam propagation method through BeampropTM software and calculations of refractive index changes in the medium with an external subroutine (Appendix B). Simulations over a wide range of intensities were performed covering from $U_0=500$ to 0.00001 .

Most of the main features observed during experimental studies in the three intensity regimes were observed in simulations. Simulations of dynamics of self-trapping in the low intensity regime, showed oscillations in beam width and intensity along the propagation direction z for a single step and over time at a fix z position. These oscillations were correlated with the existence of multiple modes in the self-written waveguide. More higher-order modes are observed at higher intensities compare to lower ones similar to experiments. An optimum self-trapping intensity was found in simulations, which is in excellent agreement with experimental results. Emergence of diffraction rings was observed during simulations at higher intensities $U_0=5$ to 0.5 . These rings presented a similar behaviour to the rings with high-order modes observed in experiments. By addition of an intensity threshold, dark rings were simulated having identical behaviour to the experimental counterpart. At even higher intensities $U_0=0.005$ to 0.001 , the formation of a single ring was observed in simulations, with an induced flattened Gaussian profile at the entrance face. At the greatest intensity of $U_0=0.00001$ the beam intensity decreases and the refractive index saturates over the transverse direction of the whole beam.

$U_0=0.00001$ Intensity maps



$U_0=0.00001$ Refractive index maps

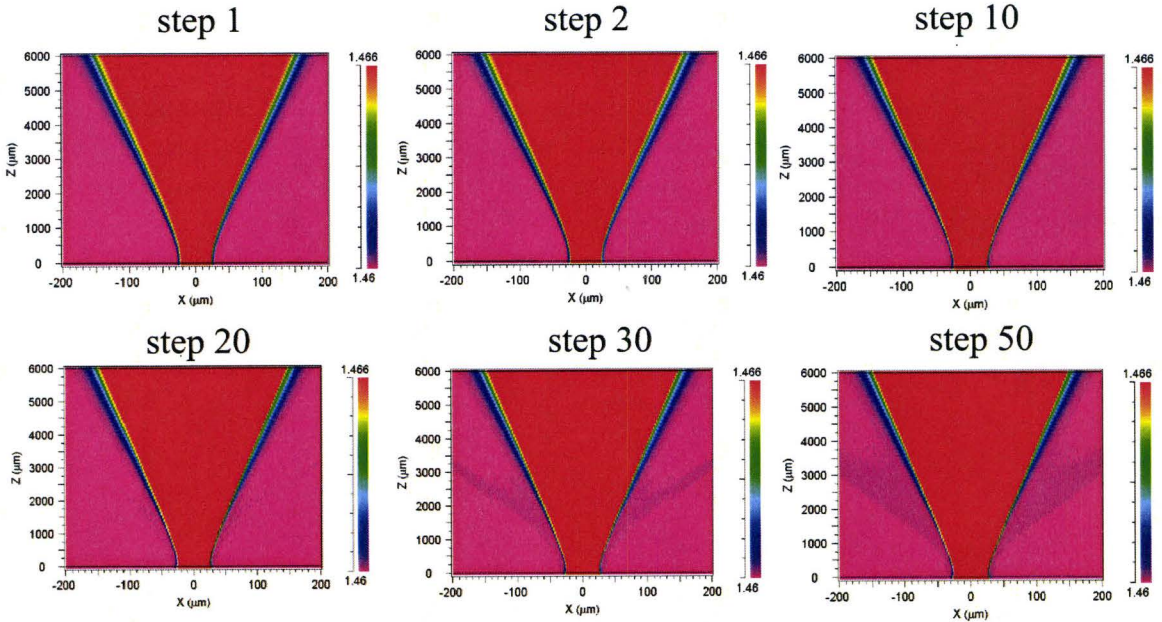


Figure 6.29: Profiles of intensity (top) and refractive index change (bottom) for $U_0 = 0.00001$ for 50 computation steps (top) obtained in Beamprop simulations. Each step represents a relative unit of time.

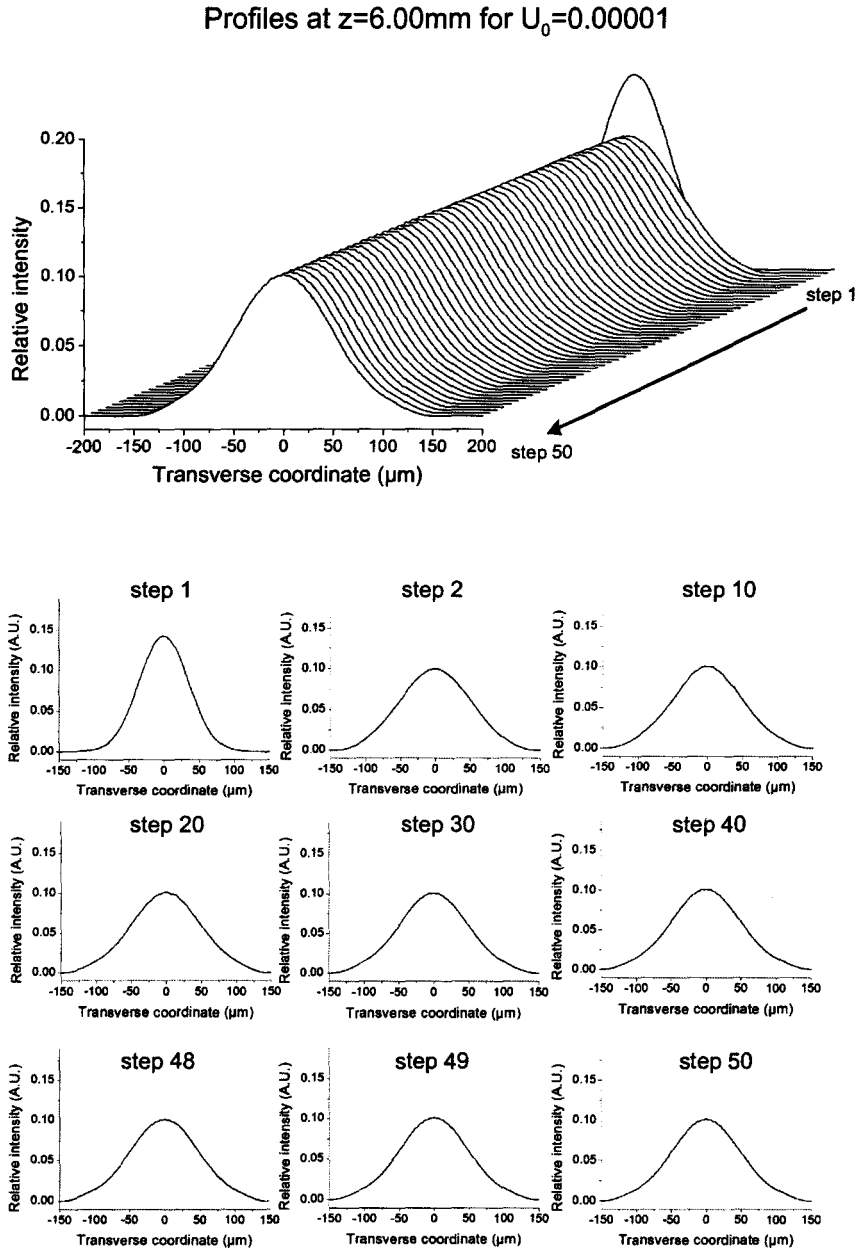


Figure 6.30: Profiles of intensity for $U_0 = 0.00001$ in the transversal direction x at the output of the medium ($z=6.00\text{ mm}$) for 50 computation steps (top) obtained in Beamprop simulations and for individual transversal profiles selected at various steps (bottom). Each step represents a relative unit of time.

Chapter 7

Conclusions and future work

The primary motivation of this thesis was to experimentally investigate laser self-action effects in a photopolymer medium covering 10 orders of magnitude in intensity, specifically the dynamics of self-trapping and nonlinear propagation effects. Experimental studies were complemented with 2D numerical simulations in order to further understand the observed phenomena. These studies represent a significant advance in the area of self-action effects in photopolymers as new self-trapping dynamics were found and novel phenomena was uncovered. The findings in this thesis contribute to the broader understanding of various spatial nonlinear phenomena in optical materials. The main contributions of these studies are described below.

We demonstrated that the use of beam profiling methods *in-situ* provides critical information of the dynamics of self-action effects in photopolymers. The slow nonlinear response (ms to s) in photopolymers allows for the use of this characterisation technique. In previous studies, the phenomena of self-trapping and filamentation had only been characterized through light scattering along the beam propagation path and the properties of the resulting self-induced structures.

Temporal evolution of laser beams in the organosiloxane was studied over a broad range of intensities covering 10 orders of magnitude (3.2×10^{-5} W/cm² to 12732 W/cm²). Four distinct forms of nonlinear light propagation were identified: self-trapping at the low intensity regime (3.2×10^{-5} W/cm² to 0.016 W/cm²), new forms of nonlinear propagation including the emergence of diffraction rings and single-ring formation at the mid (0.19 W/cm² to 16 W/cm²) and high intensity regimes (27 W/cm² to 111 W/cm²), respectively. Filamentation of the whole beam was observed at even higher intensities (159 W/cm² to 12732 W/cm²).

These findings indicated that the refractive index profile induced at different intensities in the organosiloxane, plays a fundamental role in the resulting type of nonlinear phenomenon. At low intensities the induced refractive index profile remains Gaussian maximising at a value below the saturation point. In the mid-intensity regime the induced index profile has its maximum closer to the saturation point but it remains Gaussian. At the high intensity regime, saturation of the refractive index in the axial region results in a flattened-Gaussian index profile.

In the low intensity regime, the oscillatory dynamics of the self-trapped beam was attributed to the emergence of high order modes. The formation of a multimode waveguide during self-trapping was confirmed through direct observation of the excitation of high order optical modes of the self-induced waveguide as it evolved from

single-moded to multimoded guidance. Correlation between oscillations in beam intensity and width and the modal composition during self-trapping was found. High-order modes and the fundamental mode were observed at valleys and peaks of an intensity oscillation, respectively. We found that oscillatory dynamics of self-trapped beams varied with input intensity. While oscillations in intensity and beam width were rapid and irregular at high intensities, slow and more regular oscillations occurred at low intensities. As the rate of refractive index change increased with input beam intensity, there was a corresponding increase in both the number of modes and the rate at which the modal composition of the self-written waveguide changed. These findings not only confirm previous theoretical studies in photopolymers but also provide new insights into the dynamics of self-trapping.

Monitoring the temporal evolution of the intensity profile at the output of the medium allowed to extract quantitative parameters of self-trapping including: self-focusing time, self-trapped diameter and transmittance. An optimum self-trapping intensity was found at which self-focusing time was faster, self-trapped width was smallest and transmittance (percentage of incident light that passes through the sample) was maximum. This intensity was 0.008 W/cm^2 in the organosiloxane.

In the mid-intensity regime, the dynamics of diffraction rings over distances longer than the Rayleigh range was demonstrated in the organosiloxane. Parameters such as number of rings and ring diameter were extracted over time, allowing to link polymerisation kinetics to the evolution of diffraction rings. A mechanism where significant refractive index changes occur at the entrance face of the sample and linear propagation of diffraction rings occurs through the sample was proposed. This mechanism was further explored by studying the dynamics of diffraction rings at various optical pathlengths. At short pathlengths we deduced that the propagation corresponded to near-field diffraction and at longer pathlengths to far-field diffraction. At this last pathlengths the number of diffraction rings remained the same which indicated that significant changes occur in the first 5.88 mm of the sample. At the longest pathlength, the propagation was disrupted by filamentation of diffraction rings.

Studies of the effect of beam curvature on diffraction rings dynamics were performed at long pathlengths. Rings with a dark centre were induced at $R < 0$ and rings with a bright centre were most of the times induced at $R > 0$. Dark rings induced at $R < 0$ showed filamentation over time and an increase of the dark centre diameter. Filamentation of the dark centre diffraction rings showed that the timescale at which self-trapping and diffraction rings occur, affects the susceptibility of the medium to be stable to noise. An increase in the dark centre diameter over time was found to be the result of an effective increase in beam diameter for the contribution of refractive index changes to the total phase shift. Unlike other media, the organosiloxane presents permanent refractive index changes. Structures induced by dark and bright rings were characterised after their formation which allowed for the visualization of the light propagation path.

Through careful statistical analysis of experiments, a variety of diffraction rings types were observed at $R = \infty$ including dark rings, rings with high-order modes, fingerprint rings and bright rings. The observation of rings with high-order modes and fingerprint rings was attributed to slightly lower and slightly higher pre-polymerisation of the samples, respectively. While most previous studies of diffraction rings were performed in thin samples, in our studies we investigated the propagation of diffraction rings over long distances ($\gg$ Rayleigh range), including its dependence on beam curvature and pathlength. The monitoring of the temporal evolution of diffraction rings permitted the observation of dynamics under various conditions. New types of rings were uncovered as well as propagation dynamics at various pathlengths.

Single-ring formation and its subsequent filamentation was demonstrated at the high intensity regime corresponding to intensities from 27 W/cm^2 to 111 W/cm^2 . Filamentation of the ring occurred as a result of modulation instability of the beam due to noise imposed in the system. An overall increase in the size of the single-ring and on the number of filaments was found by increasing the input intensity. Circular arrays of permanently inscribed thin waveguides were obtained as a result of the filamentation process along the single ring.

Simulations over a wide intensity range confirmed the observation of the various nonlinear phenomena including oscillations during self-trapping, diffraction rings and single ring formation. Simulations confirmed the evolution of the refractive index profile from Gaussian to flattened-Gaussian for increasing input intensity. Numerical simulations performed at low intensities confirmed the existence of an optimum intensity for self-trapping.

To summarize, the new contributions made by the studies in this thesis to the spatial self-action effects field include: the systematic intensity dependent study across 10 orders of magnitude in intensity uncovering three main intensity regimes, the direct observation of high order modes during self-trapping confirming previous theoretical studies, novel phenomena found in photopolymers including the emergence of diffraction rings and the formation of a single ring and its further filamentation and finally the key role of the induced refractive index profile at different intensities resulting in a variety of phenomena.

The work presented in this thesis is a strong base on which further work can be developed. We propose below other studies that could be carried out to better understand the self-action effects in photopolymers and to probe new phenomena.

- I. Polarization dependent studies can be performed at all intensities to observe whether the direction of the electric field of the input beam has an impact on the nonlinear light propagation in photopolymers. Previous studies have found for instance that circularly-polarized beams offer the most stable beam configuration against filamentation for Kerr nonlinearities [102]. We could investigate the polarization effects at the three intensity regimes we found. In the low and mid intensity regimes the effect of polarization on the modal properties of the

self-trapped beams and on the formation of diffraction rings can be investigated. At the high intensity regime, the effect of polarization on changes in the modulation instability dynamics of the single ring and whole beam can be explored through characterisation of the resulting filamentation.

- II. Interactions of self-trapped beams can be investigated. Previously the interaction of two self-trapped beams in urethane acrylate based monomers was examined by Shoji and coworkers [57] by looking at the angle dependence between the two incident beams. However, no information about the modal evolution through the interaction process was provided. This could be investigated by monitoring the output of two self-trapped beams and their interactions *in situ*. These interactions can be extended to the different intensity regimes found in our study and hence to different laser self-action phenomena including diffraction rings, single ring formation and filamentation.
- III. Multibeam self-trapping could have potential applications for photonic crystal fabrication as micromoulds or could be used to create nonlinear photonic crystals to study their properties to localize modes as it is currently the study in other optical media [9]. We have shown in preliminary studies that self-trapping of multiple beams can occur in the organosiloxane (see Figure 7.1). A central beam surrounded with 6 beams, was input in the medium (Figure 7.1a). During the self-trapping experiment, we observed at 1 s the diffracted form of the beam at the output face (Figure 7.1c), similar to its diffraction in air (Figure 7.1b). With time, the beam self-trapped at 131 s, showing the input flower shape. Experiments to investigate interactions between beams can be designed from this example by changing the spacing between the central beam and surrounding beams. In addition, increasing the number of input beams can be investigated to explore the self-trapping dynamics of an array of beams. This could easily be performed as a printed transparency serves as the mask for multiple beam generation (Section 2.3.2). Intensity dependent studies can be carried out with multibeams in order to observe the interactions of the different observed phenomena including diffraction rings, single ring formation and filamentation.
- IV. Permanent structures induced through self-action effects in photopolymers can be employed for different optical applications. Beams with an intensity profile similar to a doughnut have been employed in super-resolving microscopy [91]. The structure induced by the single ring intensity beam in our studies could served as an optical component to generate such a doughnut beam. To investigate the feasibility of this application, new materials that are stable after photopolymerisation could be studied, for example photopolymers using thermal initiators. Although the prepolymerization of this type of materials might leave a smaller refractive index change, it might be enough to elicit some of the self-action effects observed in this study.

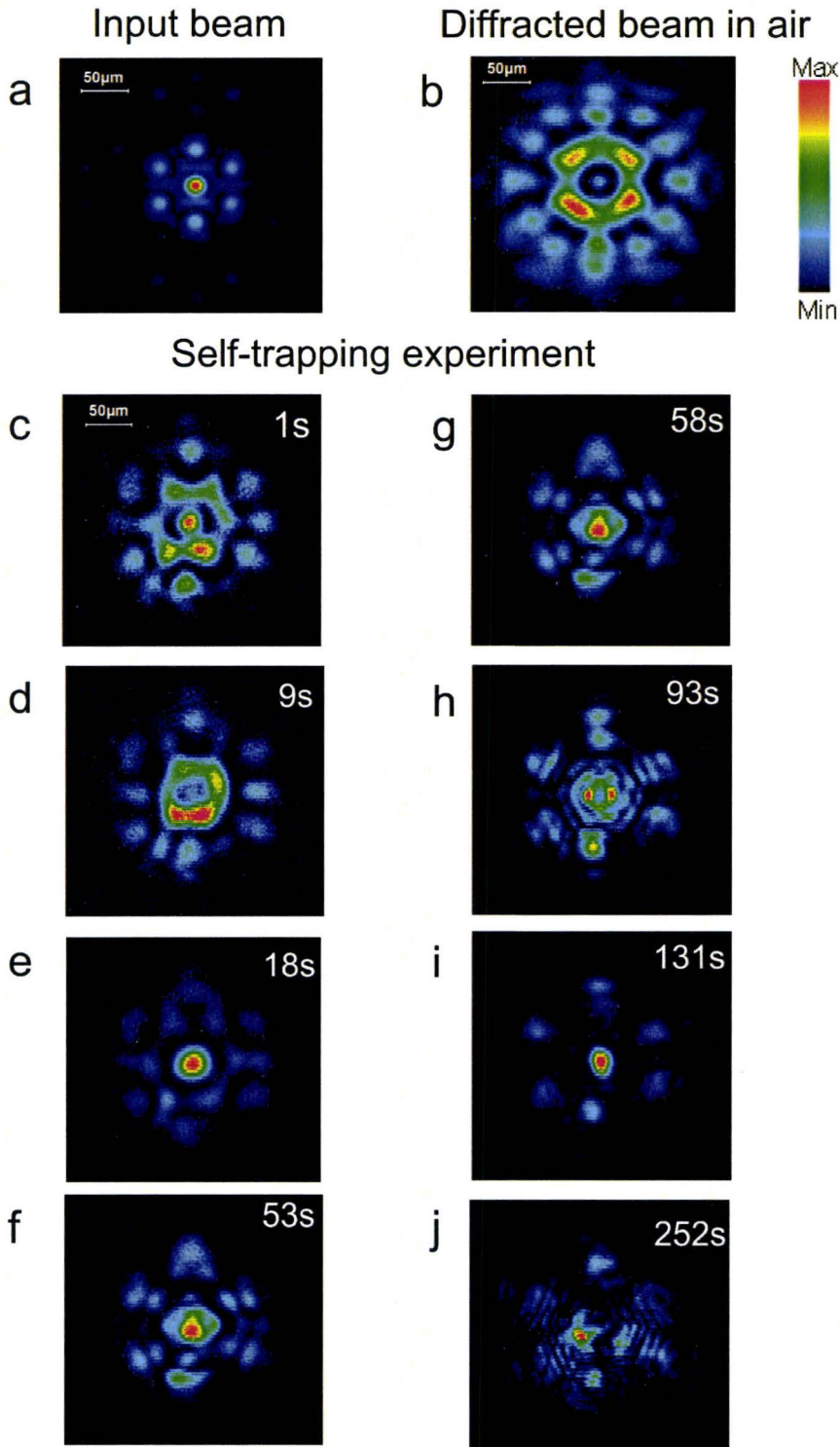


Figure 7.1: Multibeam self-trapping experiment at 0.016 W/cm^2 . 2-D intensity profiles of preliminary results are shown. The a) input beam has a central beam surrounded by 6 beams and the intensity profile after diffraction in air is shown in b). The temporal evolution of self-trapping of the multibeam is shown from c) to j) at the output face of the organosiloxane.

- V. The investigation of self-action effects in other type of materials can be explored, for example in polymer-dispersed liquid crystals. In previous studies, by using holographic techniques, gratings have been formed through the rapid photopolymerisation of the monomers in regions of high intensity leading to segregation of liquid crystals to the low intensity regions [133]. Polymer-dispersed liquid crystals combine the properties of optical anisotropy and being switchable in external electric fields from liquid crystals with the property of changing the refractive index upon illumination from photopolymers. By using the self-action effects observed in photopolymers, 3D active structures could be built.
- VI. In previous theoretical studies [35; 124; 109] and in this thesis, 2D simulations have been performed for self-trapping and other nonlinear propagation phenomena occurring in photopolymers. Numerical simulations in 3D could be carried out to have a direct comparison with experimental results. This will determine up to what degree radial symmetry can be applied in 2D simulations. In order to simulate filamentation, noise can be introduced either in the input beam or in the medium. Different types of noise including amplitude or phase, radially symmetric and linear can be probed to observe the stability of the system.
- VII. In previous studies, the refractive index change and thickness have been measured as a function of time [134] using a double-interferometer technique. The investigation of physical parameters in our system such as the refractive index change with exposure and critical exposure required to initiate polymerization (U_0) would be useful in order to correlate experimental and simulated studies more accurately. This values could be obtained by performing interferometric measurements similar to previous studies where a sample is placed in one arm of an interferometer and is uniformly irradiated *in situ* recording changes in the interference pattern over time.

Appendix A

Filamentation at different input intensities

Experimental results showing the temporal evolution of 2D intensity profiles for input intensities (powers) corresponding to: $27\text{W}/\text{cm}^2$ ($85\mu\text{W}$), $40\text{W}/\text{cm}^2$ ($125\mu\text{W}$), $64\text{W}/\text{cm}^2$ ($200\mu\text{W}$), $95\text{W}/\text{cm}^2$ ($300\mu\text{W}$) and $111\text{W}/\text{cm}^2$ ($350\mu\text{W}$) are shown below.

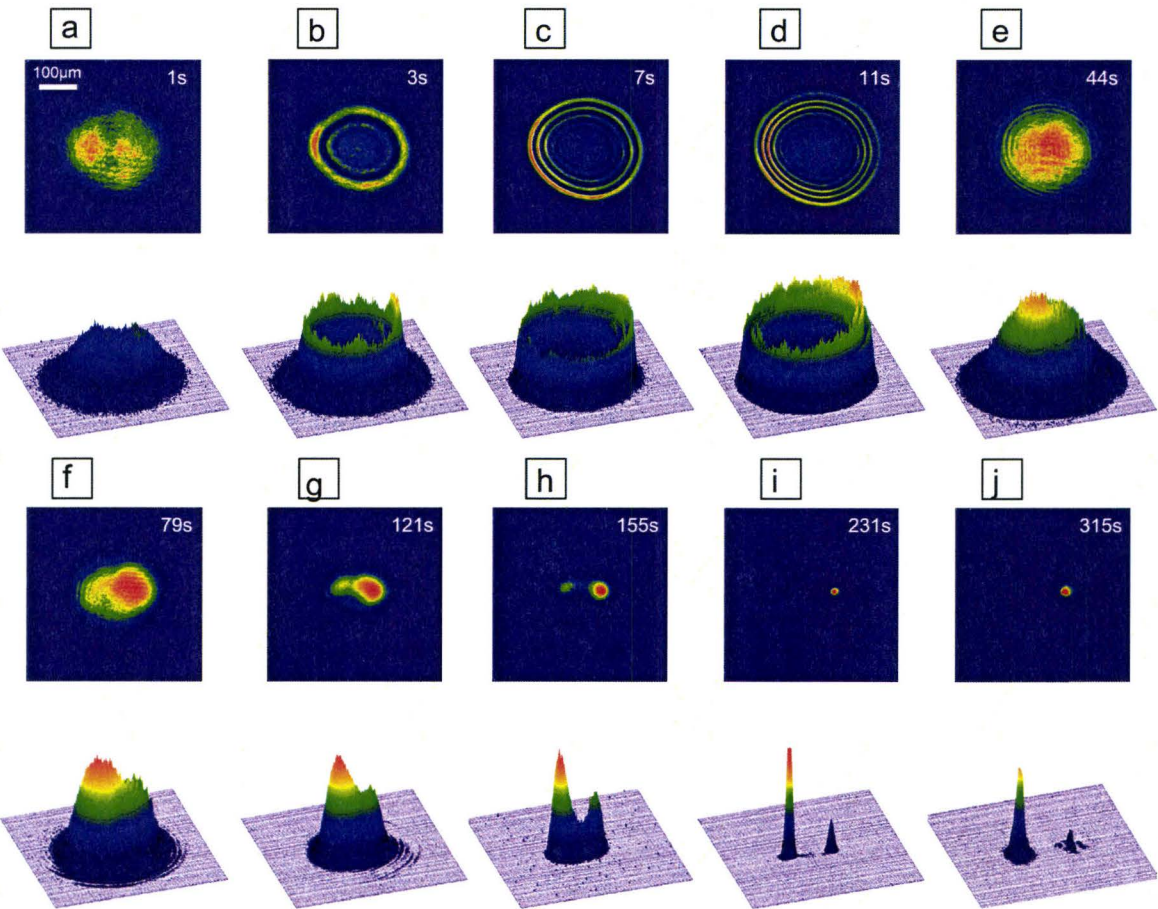


Figure A.1: 2D intensity profiles imaged at the exit face (6mm) of the organosiloxane showing the temporal evolution of an input Gaussian beam with intensity of $27\text{W}/\text{cm}^2$ (power $85\mu\text{m}$)

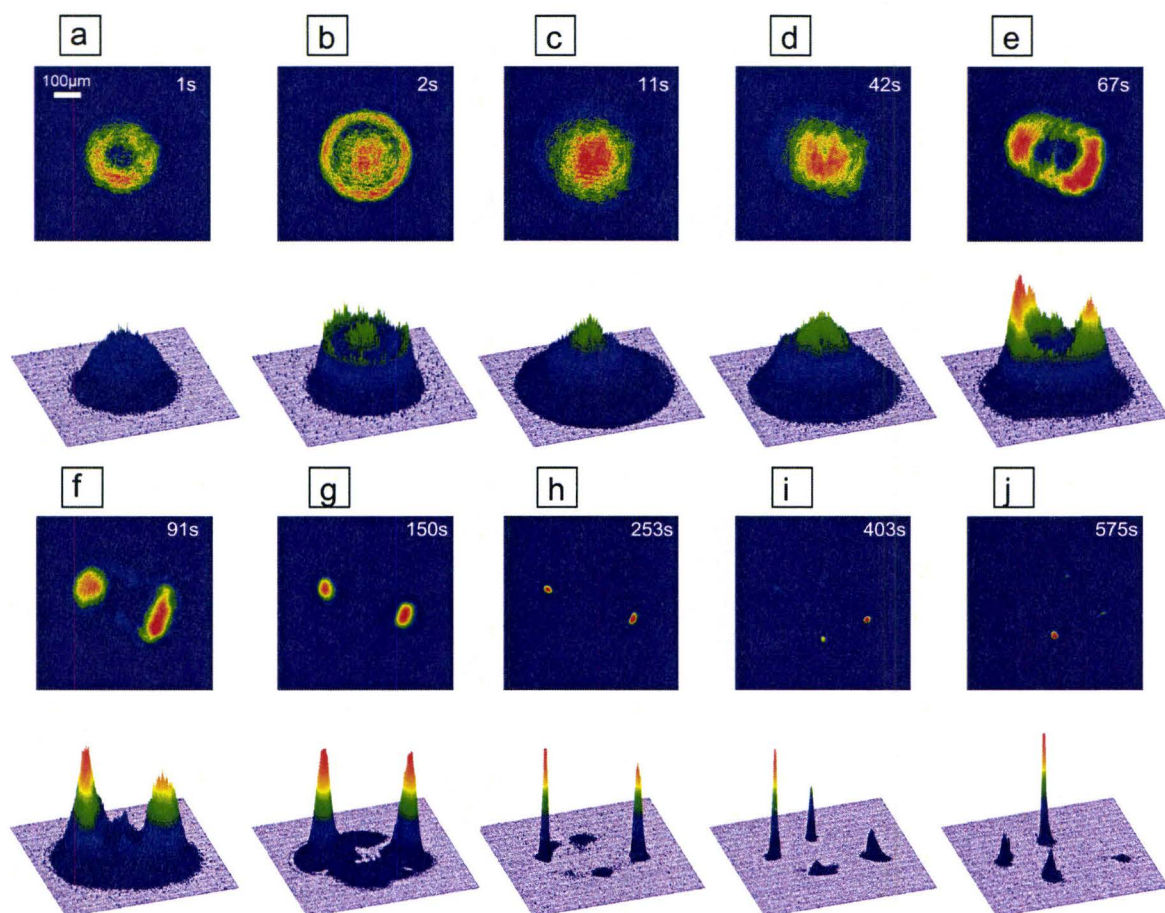


Figure A.2: 2D intensity profiles imaged at the exit face (6mm) of the organosiloxane showing the temporal evolution of an input Gaussian beam with intensity of $40W/cm^2$ (power $125\mu m$)

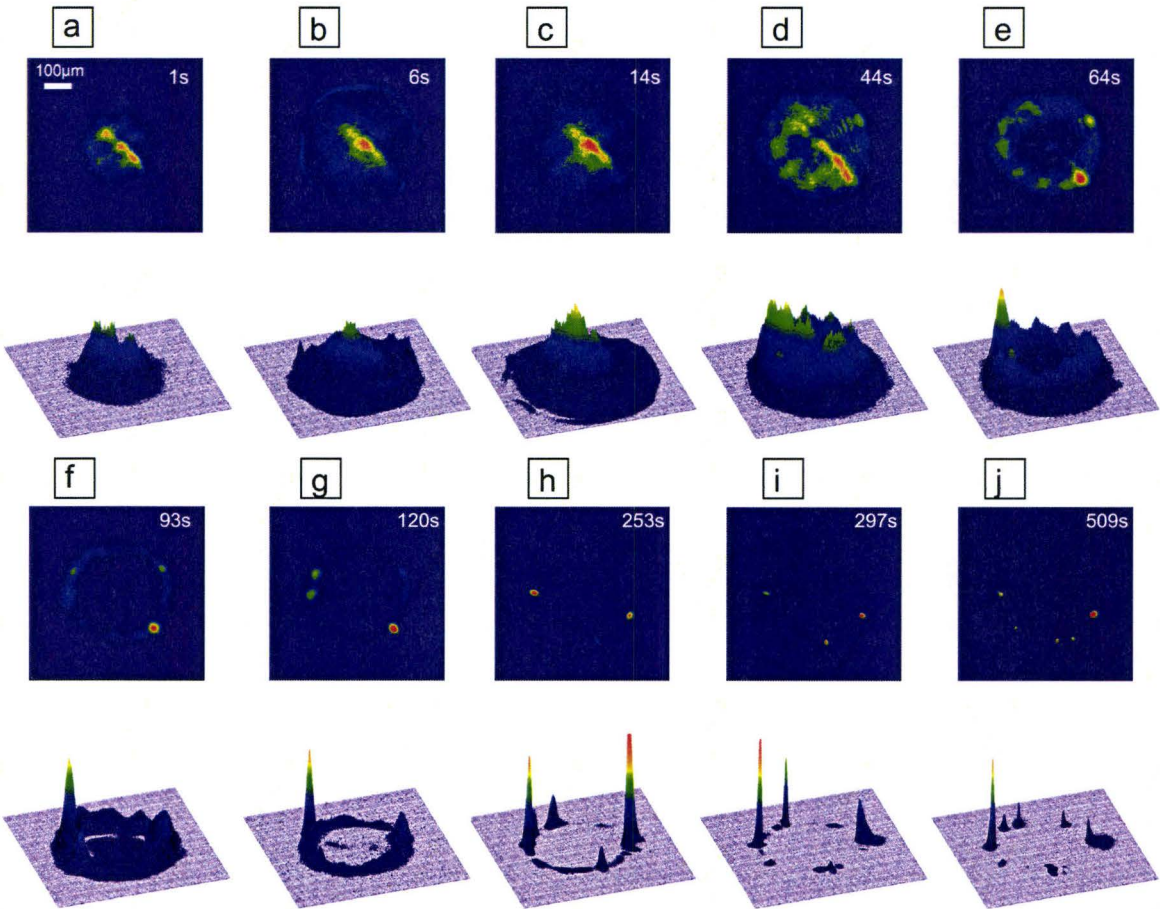


Figure A.3: 2D intensity profiles imaged at the exit face (6mm) of the organosiloxane showing the temporal evolution of an input Gaussian beam with intensity of $64\text{W}/\text{cm}^2$ (power $200\mu\text{m}$)

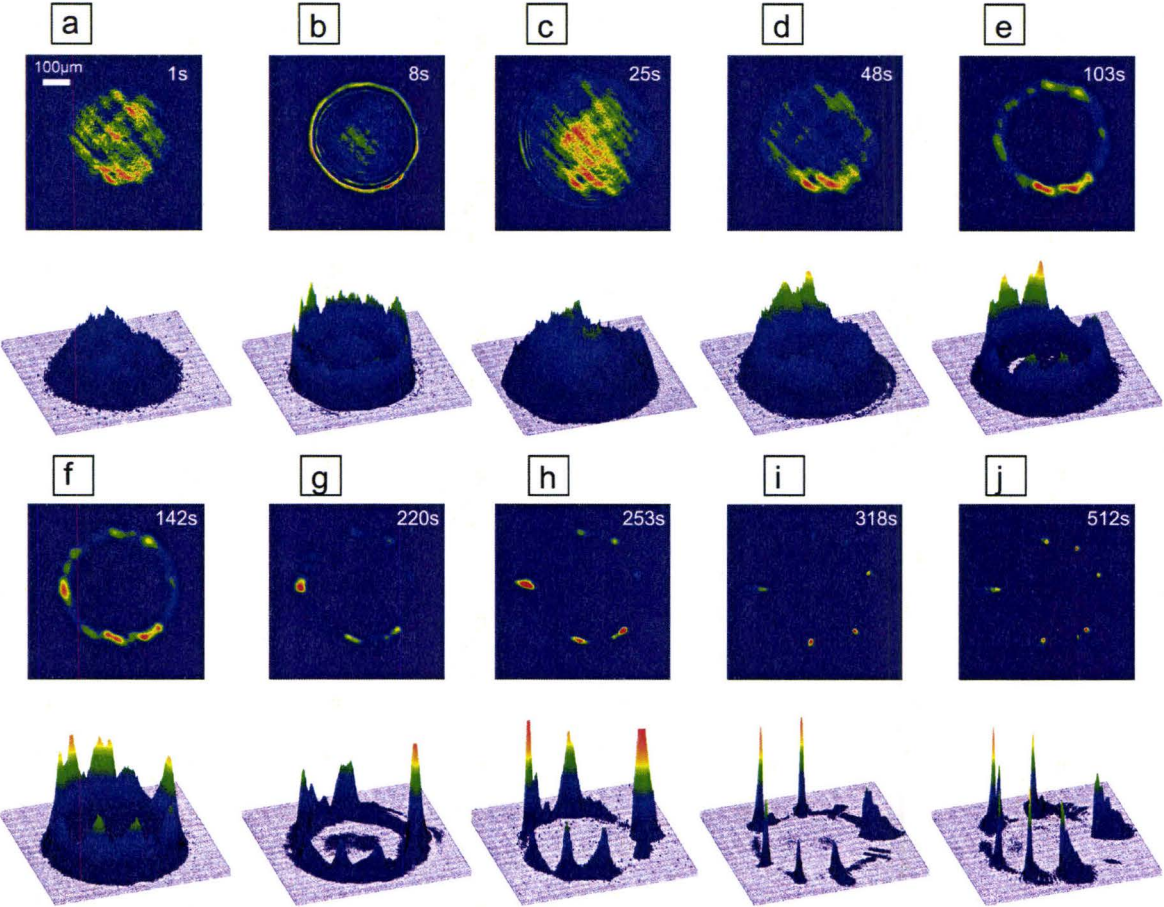


Figure A.4: 2D intensity profiles imaged at the exit face (6mm) of the organosiloxane showing the temporal evolution of an input Gaussian beam with intensity of $95\text{W}/\text{cm}^2$ (power $300\mu\text{m}$)

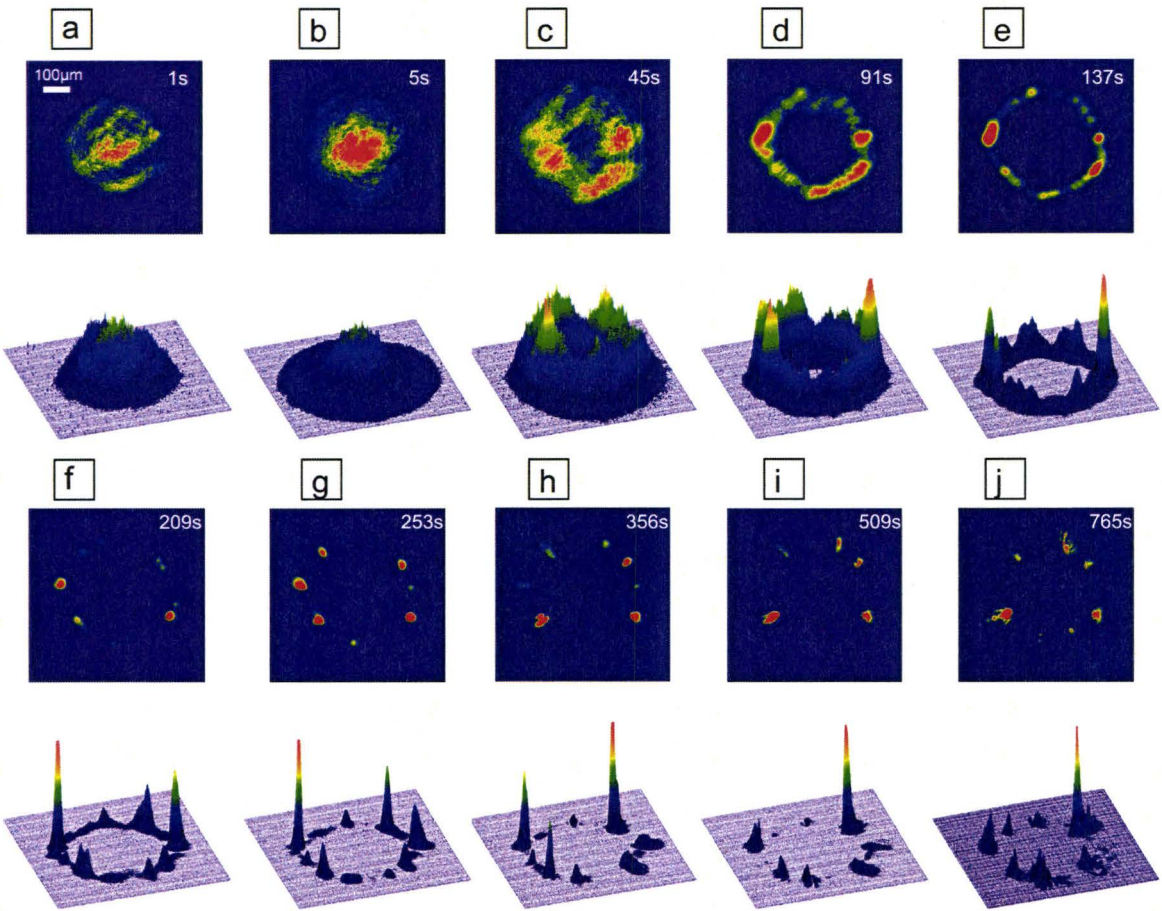


Figure A.5: 2D intensity profiles imaged at the exit face (6mm) of the organosiloxane showing the temporal evolution of an input Gaussian beam with intensity of $111\text{W}/\text{cm}^2$ (power $350\mu\text{m}$)

Appendix B

External subroutine for nonlinear propagation simulations

The external subroutine employed for nonlinear propagation simulations was used to convert maps of electric field amplitude values into maps of refractive index which were feeded into the BeamPROPTM software from RSoft Design Group, Inc. This program was initially developed by Jonathan Lannan. The program was compiled with Borland C++ compiler version 5.5. The code is shown below, all text after // corresponds to comments in the program.

```
//SIMULATION SETTINGS: Slice Step 0.4 in x, 100 in z.

//Include Header files
#include < stdio.h >
#include < math.h >
#include < stdlib.h >
#include < string.h >

//Define Variables for Simulation
#define Nx 2001          //Number of x data points per slice
#define Nz 60           //Number of slices
#define max_n 0.006     //Maximum index change of material
#define Uo 0.05         //Critical Exposure
#define thresh 0.5      //Threshold exposure

void main(int argc, char *argv[ ])
{ //Define Variables
int i,j,k, count, length, length2;
double sample, data[Nx][Nz+1];
char y[20], filename[80], step_intensity[80], index[80];

FILE *input, *out;

//take given file name and create intensity and index file names

strcpy(filename, argv[1]);
//strcpy(intensity, argv[2]);

strcpy(step_intensity, filename);
strcpy(index, filename);
```



```

length = strlen(filename) -1;

step_intensity[length-3] = '\0';
strcat(step_intensity, "_intensity.txt");

index[length-3] = '\0';
strcat(index, "_index.txt");

//Part 1: Read in Slice Values
printf("\nReading in values...");
for (j = 0; j < Nz; j++)
{if (j < 10)
    { itoa(j, &filename[length], 10);}
if(j >= 10 && j < 100)
    { itoa(j, &filename[length-1], 10);}
if(j >= 100)
    { itoa(j, &filename[length-2], 10);}
input = fopen(filename,"r");

    //Clear Header of file
    i = 0;
    fscanf(input, "%s", &y);
    fscanf(input, "%s", &y);
    fscanf(input, "%s", &y);
    fscanf(input, "%s", &y);
    fscanf(input, "%s", &y);
    fscanf(input, "%s", &y);
while (i < Nx)
    { fscanf(input, "%lf", &sample);
      data[i][j] = sample*sample;
      i++; }
    fclose(input); } printf("Complete\n");

//Part 2: Output Slice Values to file
printf("Outputting intensity profile...");
out = fopen(step_intensity, "w");
//print header
fprintf(out, "/rn,a,b/nx0/ls1\n/r,qa,qb\n %i -1 1 0
OUTPUT - REAL - 3D \n %i 0 1 \n", Nx, Nz);
for (i = 0; i < Nx; i++)
{   for(j = 0; j < Nz; j++)
    {   data[i][j] = data[i][j] - thresh;
        if (data[i][j] < 0){
            data[i][j] = 0; }
        fprintf(out, "%e\t", data[i][j]); }

```

```

    fprintf(out, "\n"); }
fclose(out);
printf("Complete\n");

//Part 3: Add to existing intensity profile
printf("Summing intensity profiles...");
if (argc==2)
{
    //printf("not skipped");
    input = fopen("c:/Ana/summed_intensity.txt", "r");          //clear header
    for (k = 0; k < 10; k++)
    {
        fscanf(input, "%s", &y);}          for (i = 0; i < Nx; i++)
    {
        for(j = 0; j < Nz; j++)
        { sample=0;
            fscanf(input, "%lf", & sample);
            data[i][j] = data[i][j] + sample; } }
fclose(input); }
printf("Complete\n");
printf("Writing new summed profile...");
//Location of the summed index file used to calculate index
out = fopen("c:/Ana/summed_ intensity.txt", "w");

//print header
fprintf(out, "/rn,a,b/nx0/ls1\n/r,qa,qb\n%i -1 1 0 OUTPUT _ REAL _ 3D \n%i 0 1\n", Nx, Nz);
for (i = 0; i < Nx; i++)
{
    for(j=0; j < Nz; j++)
    {
        fprintf(out, "%e\t", data[i][j]); }
    fprintf(out, "\n");} fclose(out);
printf("Complete\n");

//Part 4: Calculate next intensity profile
printf("Generating new index profile..."); out = fopen(index, "w");
//print header
fprintf(out, "/rn,a,b/nx0/ls1\n/r,qa,qb\n %i -1 1 0 OUTPUT_ REAL_ 3D \n %i 0 1\n", Nx, Nz);

for (i = 0; i < Nx; i++)
{
    for(j = 0; j < Nz; j++)
    {
        data[i][j] = 1.46 + max_n*(1-exp( (-1)*data[i][j]/Uo));
        fprintf(out, "%e\t", data[i][j]); }
    fprintf(out, "\n");} fclose(out);
printf("Complete\n"); printf("Generating index file..."); //Location of index profile
used by BeamProp
out = fopen("c:/Ana/index.txt", "w");
//print header

```

```

fprintf(out, "/rn,a,b/nx0/ls1\n/r,qa,qb\n %i -1 1 0 OUTPUT_ REAL_ 3D\n %i 0
1\n", Nx, Nz); for (i = 0; i < Nx; i++)
{
    for(j=0; j < Nz; j++)
    {
        fprintf(out, "%e\t", data[i][j]); }
    fprintf(out, "\n"); } fclose(out);
printf("Complete\n"); }

```

Appendix C

Nonlinear simulations in organosiloxane

Intensity and refractive index profiles for $U_0 = 100$ and $U_0 = 100$ are shown in Figures C.1 to C.4. Profiles of step 50 at various points along the propagation coordinate z are shown in Figure C.5 for various values of U_0

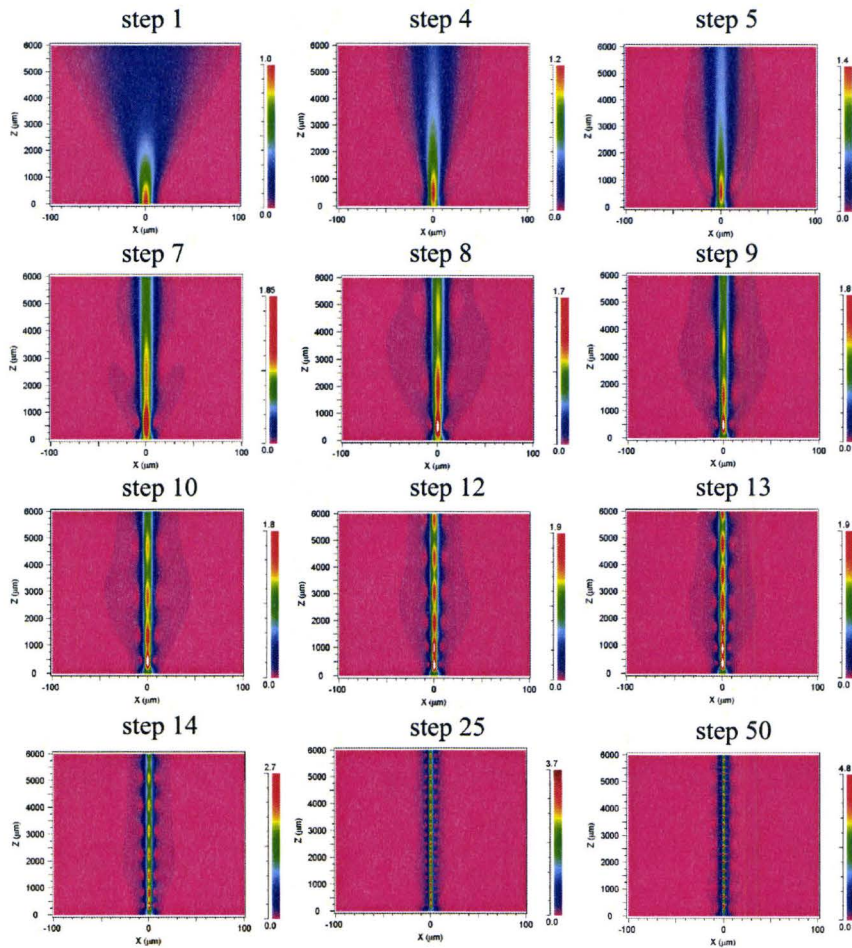


Figure C.1: 2D Simulation results in Beamprop showing the intensity profiles of a propagating Gaussian beam with $U_0 = 100$. The propagation coordinate is indicated with z and the transverse coordinate is x

$U_0=100$ Refractive index maps

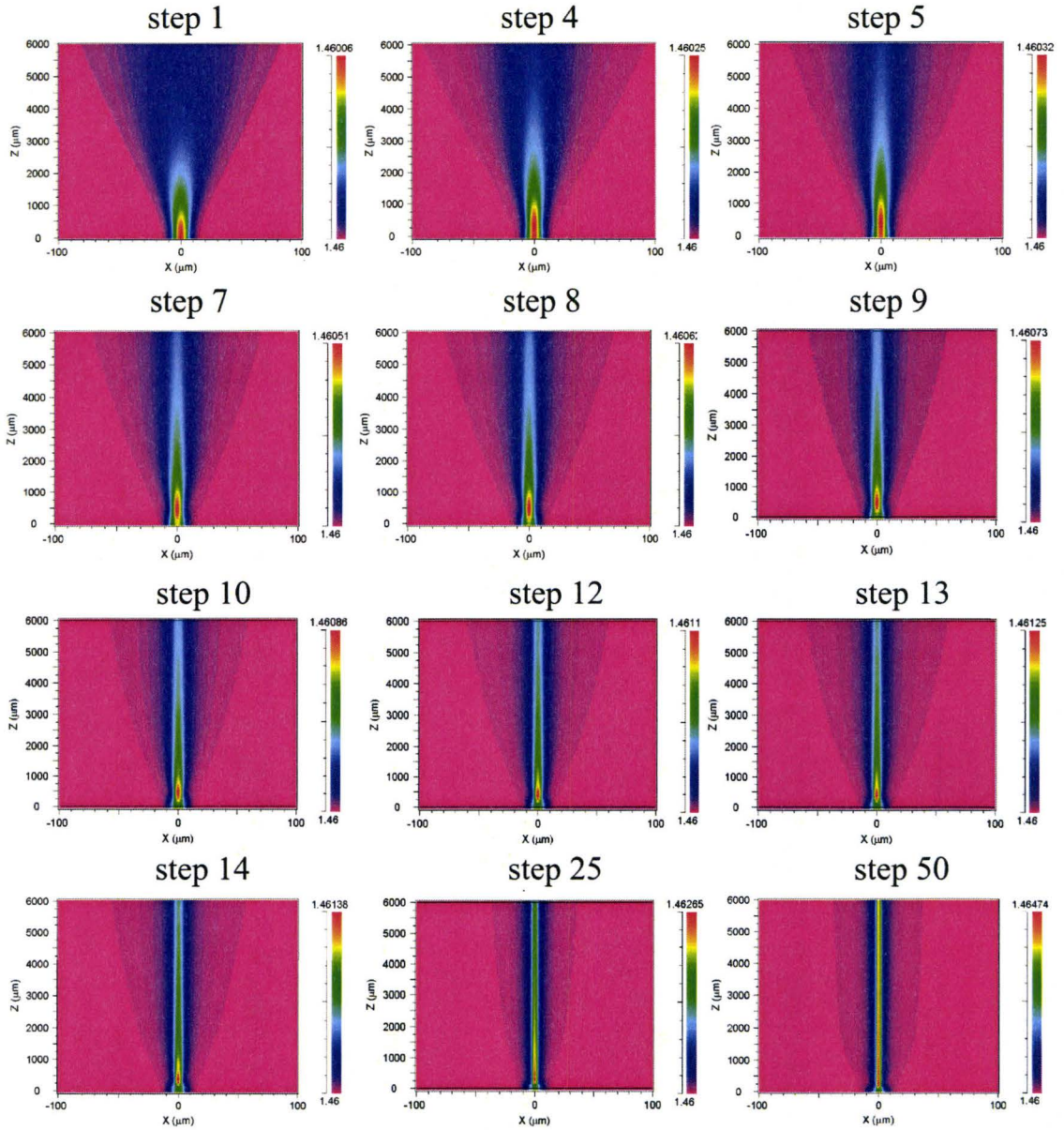


Figure C.2: 2D Simulation results in Beamprop showing the refractive index profiles of a propagating Gaussian beam with $U_0 = 100$. The propagation coordinate is indicated with z and the transverse coordinate is x

$U_0=10$ Intensity maps

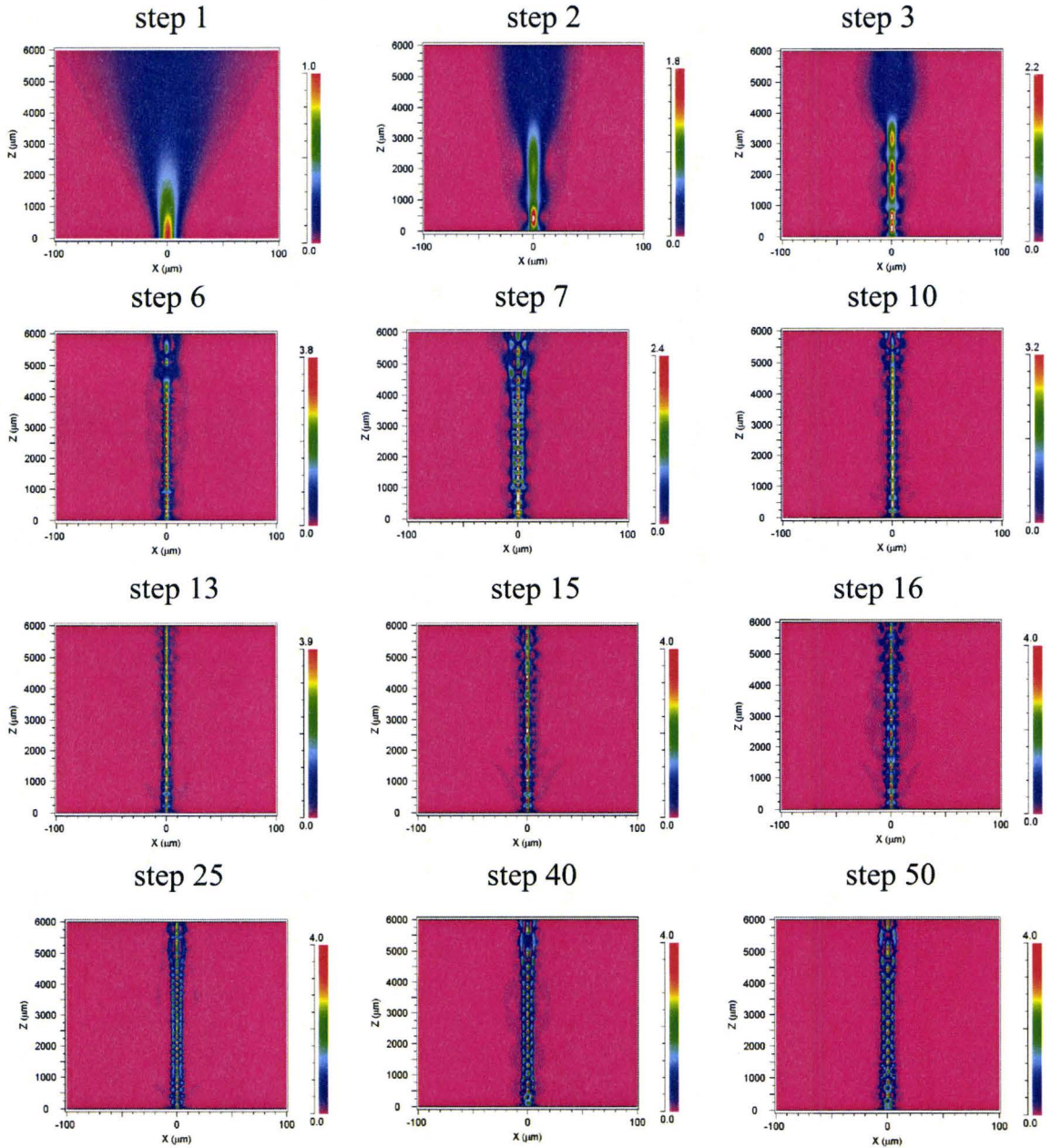


Figure C.3: 2D Simulation results in Beamprop showing the intensity profiles of a propagating Gaussian beam with $U_0 = 10$. The propagation coordinate is indicated with z and the transverse coordinate is x

$U_0=10$ Refractive index maps

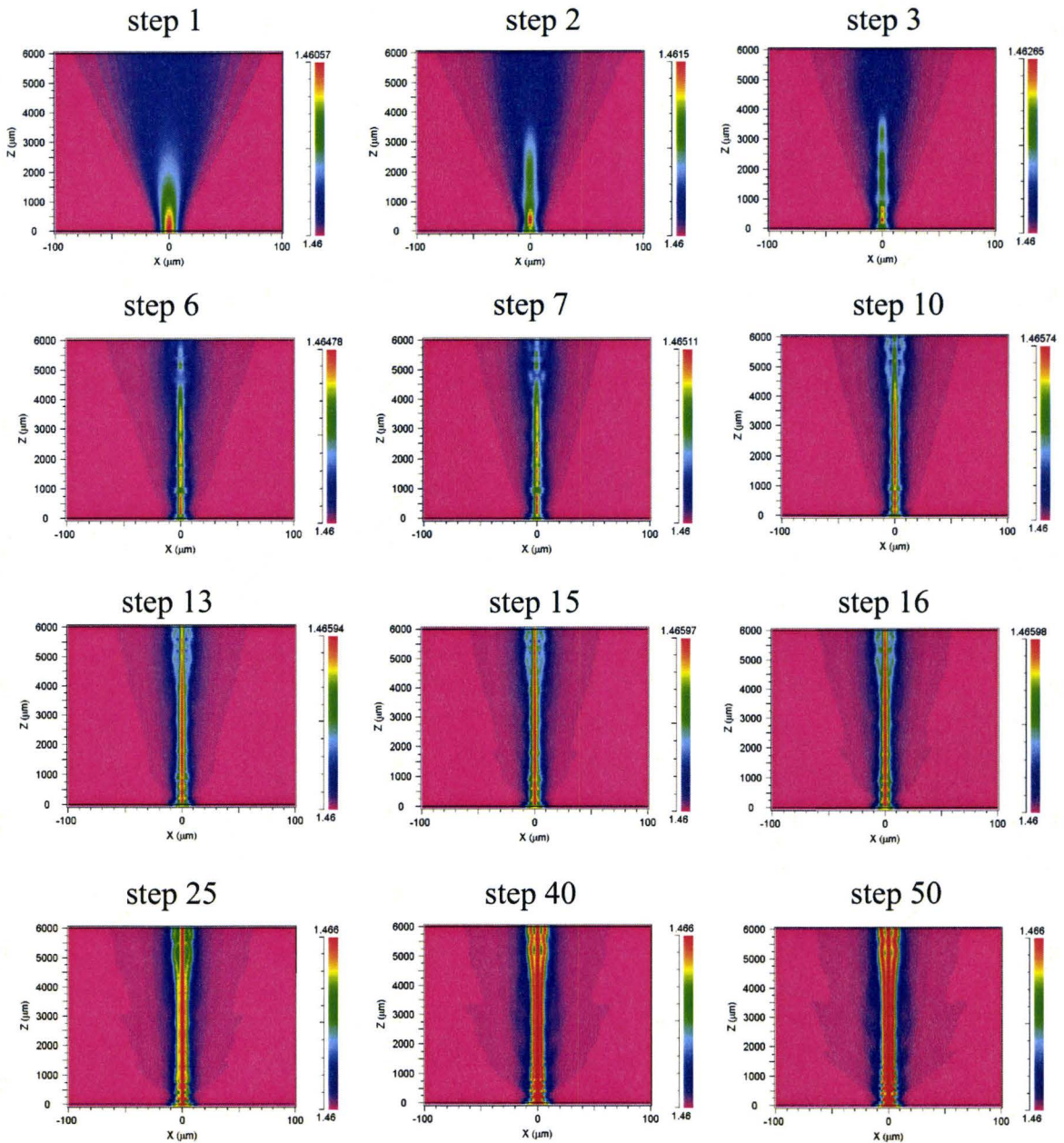


Figure C.4: 2D Simulation results in Beamprop showing the refractive index profiles of a propagating Gaussian beam with $U_0 = 10$. The propagation coordinate is indicated with z and the transverse coordinate is x

Profiles of step 50 at various "z" for various U_0

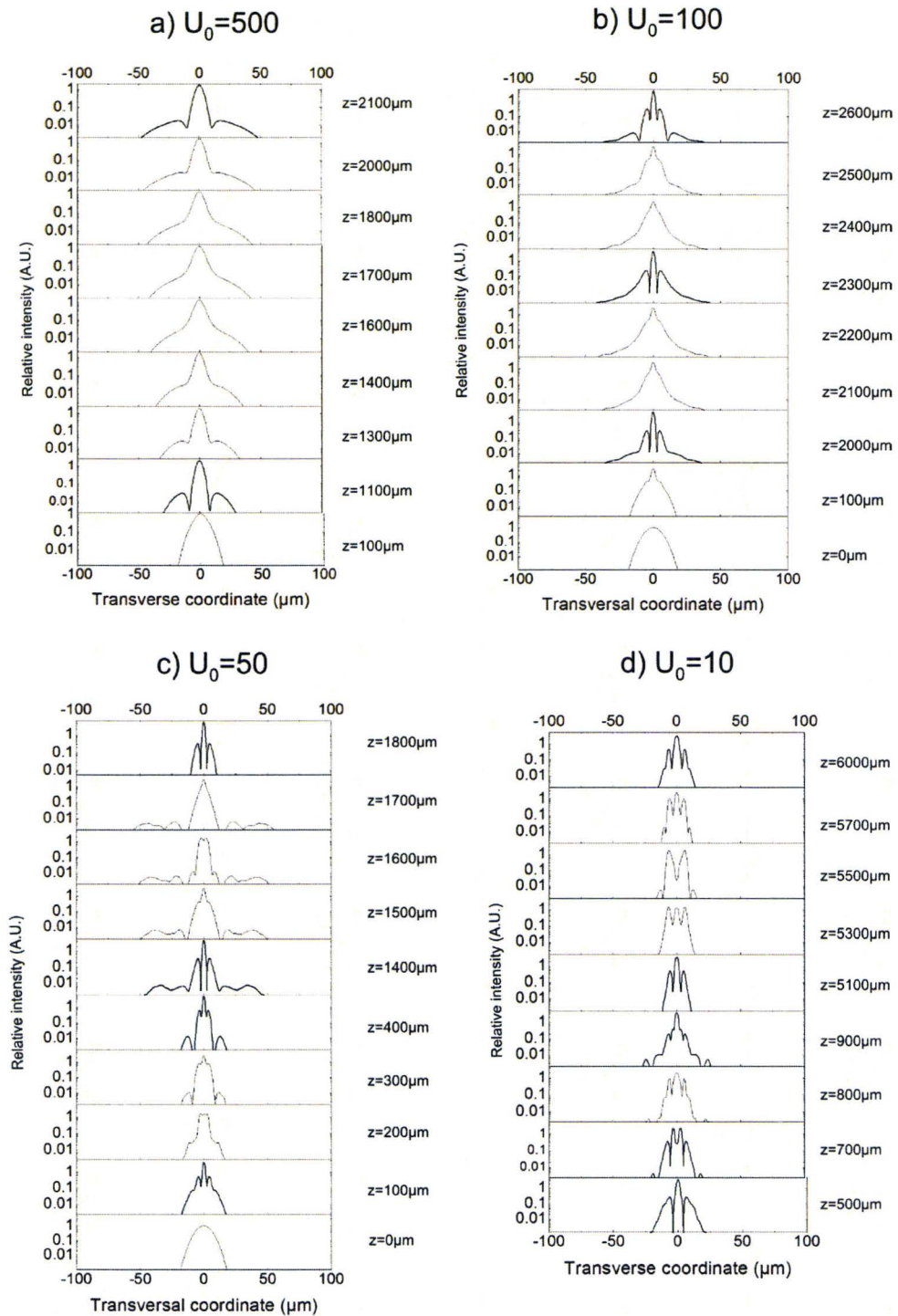


Figure C.5: Simulated intensity profiles along z for various U_0 values for step 50. Indicated in bold are profiles with intensity maxima. The relative intensity is in logarithmic scale for clarity.

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