

Light-induced self-writing effects in bulk chalcogenide glass

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Abstract – A waveguide can be self-written by a beam of light propagating in a photosensitive material. We report the first observation of self-writing effects in bulk chalcogenide glass and investigate the influences of different writing beam sizes and powers. Simulations of the process show a good agreement with the experimental results. This verifies our numerical model and allows the dynamics of this process to be explored. Using this knowledge, we predict the experimental parameters and conditions required to write waveguides, tapers and ultimately complex three-dimensional structures.

Index Terms – self-writing, waveguide, photosensitivity, chalcogenide glass, channel, taper

I. INTRODUCTION

This paper explores *self-writing* processes, in which a beam of light induces refractive index changes that dynamically evolve to form a waveguide, which then guides this light [1]. To perform self-writing, a *photosensitive* material must be used: materials are described as photosensitive if they experience a long-lasting refractive index change when exposed to light. We begin by briefly illustrating this concept, which is described in detail in Refs [1-4].

Consider a Gaussian beam focussed on a material with an initially uniform refractive index. If the material is photosensitive at the writing wavelength, this diffracting light distribution starts to change the index within the material. The largest refractive index changes occur at positions where the light intensity is highest, which in this case is along the propagation axis. If the index *increases* in response to illumination, this change acts to reduce the diffraction of the incoming beam. In these early stages, diffraction imprints an adiabatic taper within the material, and over time this taper can evolve into a channel waveguide, guiding the writing beam through the material.

This process is called self-writing since the same beam that creates the waveguide is subsequently guided by it. After it has formed, a self-written waveguide can be used to guide light at other wavelengths. As these changes in index can be permanent and local, this process is distinct from other self-action effects such as spatial and photorefractive solitons.

Waveguides are currently formed using many different techniques, including epitaxial growth, diffusion methods and direct-writing [5]. The direct-writing technique has the advantage that using photosensitivity, avoids the complex chemical processing used in the other techniques. However, in direct-writing, either the sample or the beam must be translated to pattern the waveguide. In contrast, using self-writing, the waveguide evolves dynamically, and so it is a one-step process that requires no translation. Theoretical work [2] shows that the form of the resulting waveguide can be tailored by appropriate choice of writing beam shape, which allows the formation of a wide range of complex structures. In addition, since self-written waveguides

evolve dynamically, they typically vary adiabatically, and so should experience low scattering losses. This is a relatively new area of research and in this paper only channel waveguides are considered. The evolution of self-written waveguides in bulk glasses is a rich new area of physics that has not yet been explored.

Self-writing can be realised in a variety of materials: what is required is a long-lasting change in refractive index in response to illumination. Previously, self-written channel waveguides have been created in both planar germanosilicate glass [3] and As_2S_3 chalcogenide glass [6]. This process has also been studied experimentally in the bulk geometry, where solid waveguides have been formed in a liquid photopolymer [7], tapers have been formed in UV-cured epoxy [8] and recently a three-dimensional photonic crystal was fabricated by self-writing channels in a photopolymerizable resin [9,10]. The fact that self-writing has been carried out in such a wide range of materials demonstrates that it is a robust process that can occur regardless of the precise chemical or structural mechanisms responsible for the index change.

Simulations of the self-writing process completed to date have principally considered the planar geometry, which is less computationally demanding than the bulk case. These studies have been performed both for glass [1] and liquid photopolymers [7]. Some preliminary simulations for bulk glasses have also been done, which show that a channel waveguide can form in a bulk material providing that a sufficiently large refractive index change is possible in the material [4].

In this paper we explore self-writing in bulk glass, both experimentally and theoretically. Self-writing has not previously been experimentally demonstrated in any bulk glass. The bulk geometry offers greater flexibility than the planar geometry to form complex, three-dimensional waveguides. Also, glasses offer the advantage of straightforward integration with conventional glass technologies.

II. MATERIAL

In these experiments a compound chalcogenide glass, gallium lanthanum sulphide (Ga-La-S , $n = 2.5$ [11]), is used. This family of glasses typically has the potential for large photosensitive index changes under illumination [11], although this photosensitivity is not yet well characterised. The properties of Ga-La-S can be altered somewhat by doping the glass, and in our experiments pure Ga-La-S , Ga-La-S oxide and cerium-doped Ga-La-S have been used. We find that the biggest index changes are achieved in the cerium-doped sample, and these are the results presented here. The composition of this glass is $70\text{Ga}_2\text{S}_3:27.5\text{La}_2\text{S}_3:1\text{La}_2\text{O}_3:1.5\text{Ce}_2\text{S}_3$, which corresponds to a doping concentration of 1.5 mol % cerium.

Light with a wavelength near the band edge typically induces changes in a material, as significant absorption occurs here, and for Ga-La-S the band edge is located near 500nm [12]. Therefore, one might expect that a two-photon absorption process would lead to photosensitivity around $1\mu\text{m}$. This wavelength is preferable as losses through the material are lower here than near the band edge. In our experiments a diode pumped Nd-YLF fibre laser ($1.047\mu\text{m}$) is used to illuminate the sample. For these experiments a high spatial quality of the writing beam is required and therefore fibre lasers are favourable. Also, the input and output faces of the sample are both polished in order to obtain parallel, high quality faces.

III. EXPERIMENT

In these experiments, a Gaussian beam is focused down to a waist on the input edge of a 5mm long sample using a lens. Another lens is used to image the output beam onto a beam profiler. The evolution of the index change is observed by monitoring the beam shape at the output face of the sample. Alignment is crucial in this experiment, and good care must be taken to position the waist on the sample edge, and it is important also not to pre-expose the sample to light.

Initially the writing beam diffracts freely in the uniform bulk material. If the refractive index of the glass increases in response to light, then we expect the beam at the output edge of the sample to narrow and become more intense as the light begins to be guided by the index change it induces. Fig. 1 shows this evolution for a typical experiment: the change in beam shape confirms indeed that self-writing has begun to occur in the material.

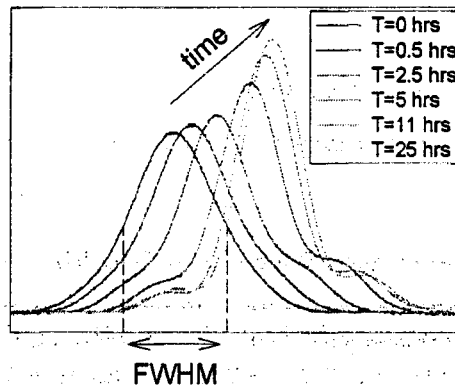


Fig. 1. The shape of the beam emerging from the sample: the beam narrows and becomes more intense during the exposure.

Fig. 2 shows the full width at half maximum (FWHM) and the peak intensity of the output beam as functions of time for two different experiments. In both these experiments a writing beam of $9\mu\text{m}$ and a power of 650mW are used. First we concentrate on Fig. 2a, which correspond to the output beams shown in Fig. 1. It can clearly be seen how the beam narrows and the peak intensity increases during the exposure (both by a factor of 1.5). During this exposure the loss does not change significantly (the power decreases by 4%).

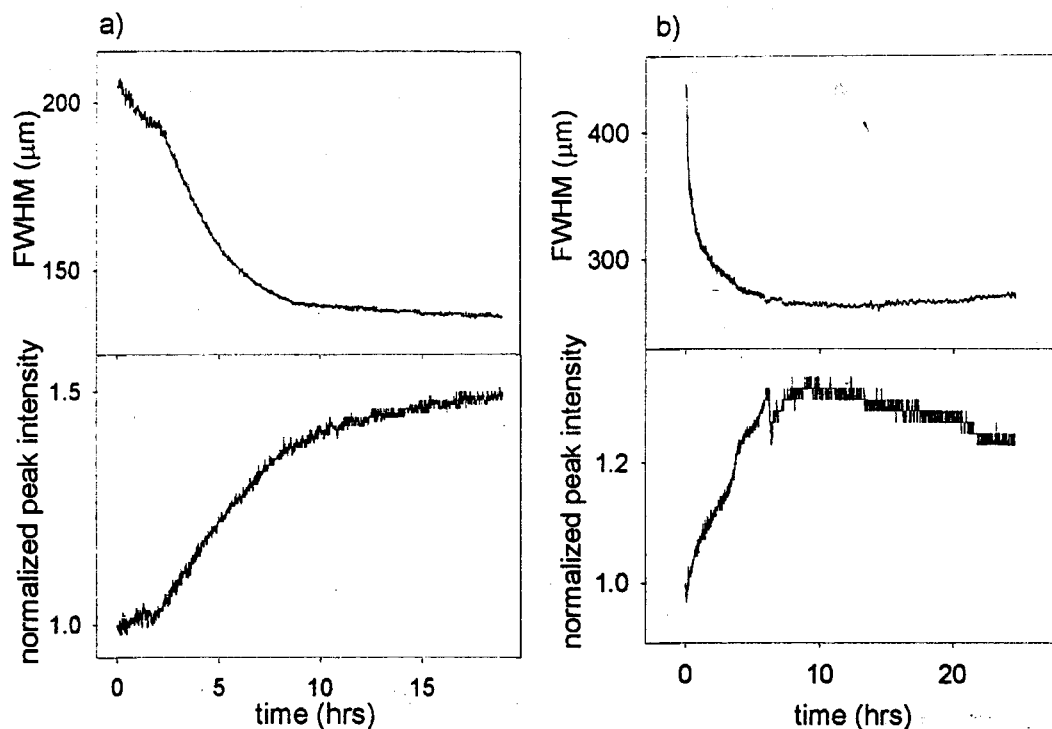


Fig. 2. The FWHM and the peak intensity of the output beam from two different experiments using a writing beam width of $9\mu\text{m}$ and a power of 650mW .

Unlike the experiment in Fig. 2a, in Fig. 2b, the FWHM and the peak intensity do not change by the same degree, and indeed the intensity even starts to decrease towards the end of the exposure. One explanation for this difference is that these samples are somewhat inhomogeneous due to crystallisation within the glass. We have confirmed the presence of inhomogeneity by performing both beam quality and loss measurements. Even for the experiment in Fig. 2a, the beam becomes slightly non-Gaussian over time as shown in Fig. 1.

Approximately 7 hrs after the start of the exposure, the FWHM levels off and stops changing. It seems likely that at this point the saturation value of the refractive index has been reached and the material cannot change any further (this behaviour is discussed later).

The longevity of this change in refractive index has been investigated by monitoring the sample a number of times after the initial exposure, as shown in Fig. 3. It can be seen that the index changes are long lasting and do not decay significantly when the light is removed, at least over the timescale of a day. However, over a longer time scale of a few days, the changes are observed to decay. In future experiments, it may be possible to make these changes permanent by annealing the glass after the initial exposure.

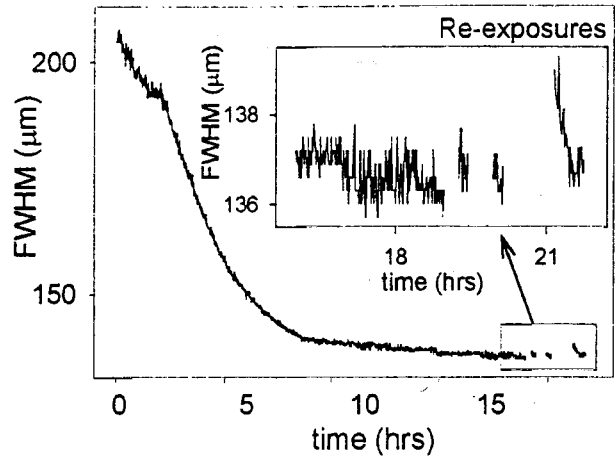


Fig. 3. Measurements of the width of the outgoing beam after the initial exposure (corresponding to Fig. 2a). The changes within the material are long-lasting.

For a given writing beam, the structure that evolves in the material is principally determined by the maximum change in refractive index that can be induced. We show that this necessitates a careful choice of the writing beam size. To obtain significant diffraction, at least a few Rayleigh ranges must fit within the material. However, very narrow beams cannot be used, since a prohibitively large refractive index change is then needed to counteract their diffraction. In the other extreme, the relatively large Rayleigh range of a wide beam results in a need for long samples. This is not only impractical for devices, but in addition losses in the material then become problematic. In practice, for any particular material, there exists an optimum choice of experimental parameters. In order to explore how self-writing depends on the writing beam width and power, a range of experiments was conducted as shown in Fig. 4. These experiments correspond to 10-20 Rayleigh ranges within the 5mm long sample.

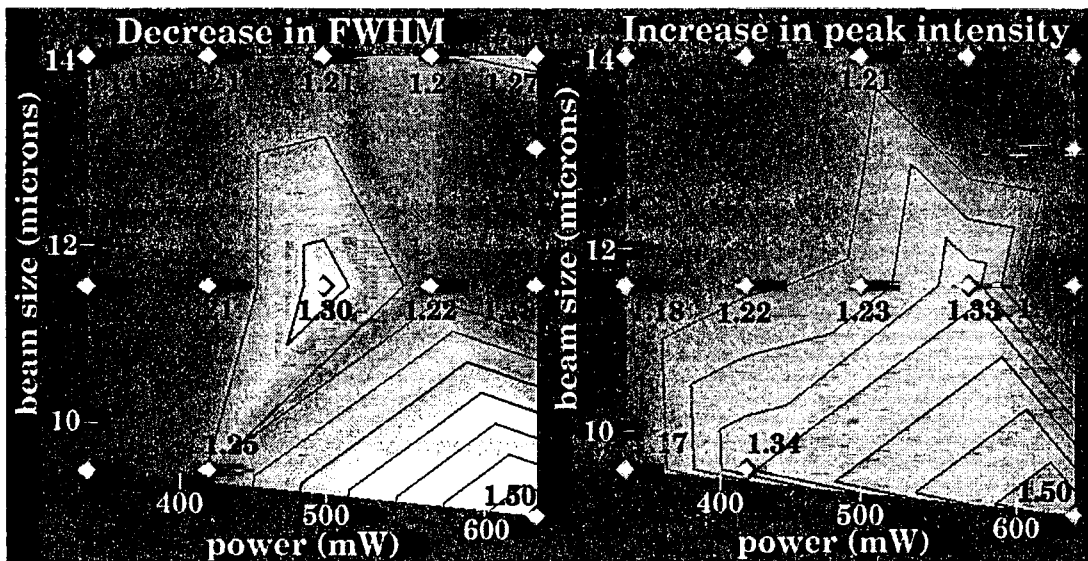


Fig. 4. Summary of experiments carried out using different writing beam sizes and powers.

We first consider the changes in FWHM. Notice that for this case the best result was achieved by the experiment shown in Fig. 2a, using a writing beam width of $9\mu\text{m}$ and a power of 650mW . It can be seen that there exists a complex relationship between the writing beam size, power, and the resulting degree to which the beam is guided by the index distribution written in the sample. Here these trends are explored.

Fig. 4 shows that narrow beams and high power together result in bigger overall changes. Hence we observe that high light intensities cause slightly greater index changes in the material that allow self-writing to proceed further. For wider beams, although bigger changes are achieved when high powers are used, the results are not as sensitive to the power as for narrower beams. Since wider beams require a higher power in order to achieve the same light intensity, in order to get more dramatic changes in this material, the power would have to be increased significantly.

Some experiments carried out using high intensity beams, i.e. $9.5\mu\text{m}$ beam and 650mW , resulted in only minor overall changes (these are not included in Fig. 4). We believe that when these high intensities see imperfections on the sample edge, in these cases damage to the edge occurs. This damage prevents a waveguide from forming, hence results in smaller changes. In conclusion, although we find that self-writing proceeds further at high intensities, in such cases damage of the sample is more likely, hence more consistent results are obtained using a slightly wider beam.

It can also be seen in Fig. 4 that the FWHM and the peak intensity do not always change by the same amount. The peak intensity is particularly dependent on the loss in light intensity in the material. Due to sample inhomogeneities the sample loss varies somewhat with position, and in addition the loss may change during the exposure, a behaviour that has been observed previously in planar glass [2]. In Fig. 4 only a selection of experiments are presented, which all showed reasonable losses in intensity, good quality beams and no damaging of the sample.

We observed that self-writing proceeded faster when the light intensity was higher (i.e. a narrow beam with high power produces the fastest change). This observation is consistent with theoretical models for photosensitivity [7,13]. However, the inhomogeneities in the sample make it hard to interpret the rate of change in refractive index.

These experiments unambiguously demonstrate self-writing effects in these glasses and help to ascertain the parameters that are most critical in this process. In the experiments carried out here a typical change in FWHM is of a factor of 1.5, when using a $9\mu\text{m}$ wide beam in a 5mm long sample. However, to form a mature channel waveguide, much larger changes are typically expected (see section V). Next we use simulations in order to predict how to increase the magnitude of the self-writing effect in bulk glass.

IV. SIMULATIONS

In order to obtain a deeper understanding of the dynamics of the self-written evolution and to predict the effects of using different experimental parameters, we have conducted numerical simulations of this process. Two differential equations can be

used to describe self-writing [1]; the paraxial wave equation describes the propagation of light through the material:

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

while the photosensitivity equation describes the refractive index evolution:

$$\frac{\partial \Delta n}{\partial T} = I^p \left(1 - \frac{\Delta n}{\Delta n_s} \right). \quad (2)$$

Here $E = E(x, y, z, t)$ is the electric field envelope amplitude, z is the propagation direction, x and y are the transverse coordinates, n_0 the initial refractive index, $k_0 = 2\pi/\lambda$ is the wave number and the loss in decibels per unit length is 4.343α . The intensity, I , is defined as EE^* and p is the number of photons involved in the process (typically 1 or 2). Here T is a normalized time defined as

$$T = a^2 k_0^2 n_0 A_p I^p t \quad (3)$$

where A_p is a real coefficient that depends on p , the material properties and λ , and t is the time in seconds.

In equation (2) a saturation index, Δn_s , is introduced to limit the change in index that can occur, and in this model the index change slows down exponentially as it approaches this value. This simple physical model has been shown to agree well with experiments in photopolymers [7] and germanosilicate glass [3]. It is also consistent with our observation that the process proceeds faster at high light intensities. However, this simple model cannot be completely accurate since we observed that the degree of index change is somewhat intensity dependent. In bulk geometry, Δn_s is crucial since the incoming light is focused in two transverse directions, resulting in a high intensity and hence a larger change in refractive index. Indeed, it has been shown previously that a saturation index must be included to prevent catastrophic collapse of the beam [4].

A Gaussian beam is used as the writing beam and so the initial condition at the input face ($z = 0$) is

$$E(x, y, 0, t) = E_0 \exp\left(-\frac{x^2 + y^2}{a^2}\right). \quad (4)$$

The beam width at the input face is a , which corresponds to a FWHM in the intensity of $a\sqrt{2 \ln 2}$.

The propagation of light through a material must in general be described using the vector wave equation [14]. However, if the beam properties vary only slowly in the propagation direction, the paraxial approximation can be used, as given by equation (1). The Gaussian beam considered here propagates along the z -axis, and so this approximation is valid so long as the diffraction of the beam is not too large. In other

words, there exists a minimum beam size, $a \gg \sqrt{2}/(k_0 n_0)$, which is always satisfied here.

To solve the equations describing the self-writing process, advantage is taken of the difference in time-scales for the processes involved. Typically the light propagation occurs in nanoseconds whereas the evolution of the refractive index takes minutes or hours. Therefore equation (1) and (2) can be treated as two independent processes that can be solved separately. This is done numerically on a grid inside the material using a split step beam propagation model. Note that for the bulk geometry these calculations are computationally intensive, and no simulations have previously been done using real parameters and conditions these compound glasses. Typical calculations use grids ranging from $[128 \times 128 \times 500]$ to $[256 \times 256 \times 600]$, which requires about 0.1 – 2 Gbytes computer memory, and these calculations take between days and weeks to run on a 1GHz processor.

To compare the simulation results with experimental data, the time in the simulation must be scaled, since a normalized time (T) is used in the model and the parameter A_p is unknown (recall equation (3)). In these materials, the index change that can be achieved by illumination (Δn_s) depends critically on the glass composition, and is typically in the range 0.0001 - 0.001 [15]. Therefore, within this range we choose Δn_s to fit the experimental data.

In Fig. 5 experimental results, from Fig. 2b, using a writing beam size of $9\mu\text{m}$ are shown together with the results from the corresponding simulation. This experiment was chosen since it demonstrates a smooth change in FWHM, which should make it easier to compare with theory. In this particular simulation we obtain good agreement for the shape of the width evolution when Δn_s is 2.5×10^{-4} , which is a physically reasonable value. It can be seen that the experimental result and the simulation start to differ somewhat at later times, which is caused by either inhomogeneities in the sample, or our simplified model for the index change, and these discrepancies accumulate over time.

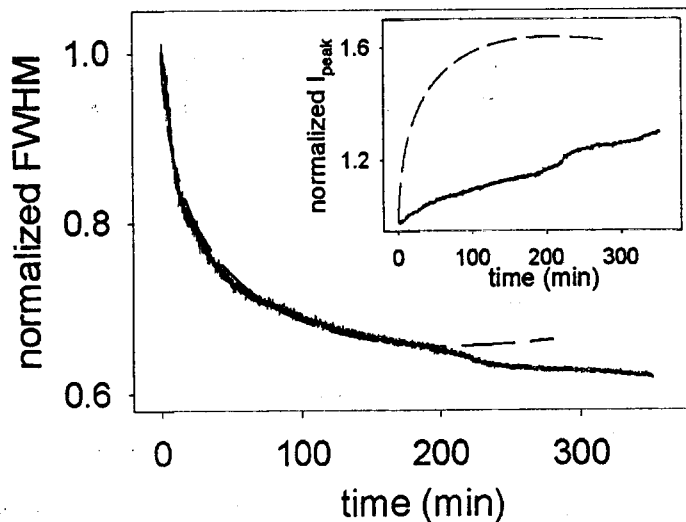


Fig. 5. Change in FWHM for experimental data (FWHM= $9\mu\text{m}$, solid lines) and simulation using a Δn_s of 2.5×10^{-4} . The inset shows the corresponding changes in peak intensity.

The inset in Fig. 5 shows that, although the simulation accurately predicts the behaviour of the FWHM, the change in peak intensity differs significantly between experiment and simulation. As mentioned earlier, it has previously been shown that loss has greater influence on the peak intensity than on the FWHM [16] and so simulations including loss were undertaken. Initially the simplest case of constant loss was considered, but agreement with observations could not be improved significantly. Next the model was extended to include a variable exposure-dependent loss, in which the local value of the loss increases in response to changes in the refractive index, as in Ref [16]. In our case, cerium-doped Ga-La-S is inhomogeneous, and so the loss differs for different writing beam locations. Therefore, the magnitude of loss needs to be fitted separately for each experiment and here the experiment shown in Fig. 5 is considered.

Fig. 6 shows again the experimental and simulation data from Fig. 5, together with results from a simulation including this variable loss, represented by solid, dashed and dotted lines respectively. The best agreement was found when using a variable loss of $1.6 \times 10^5 \Delta n$ dB/cm, hence the greatest loss which occurs in this case is 16dB/cm and will occur at localised positions where the saturation value of the refractive index has been reached. It can be seen that using this variable loss, the magnitude change in intensity can be matched well with observations, although the shape of the curves differ. It can also be seen in the inset that although the change in FWHM is affected slightly by the introduced loss, it remains good.

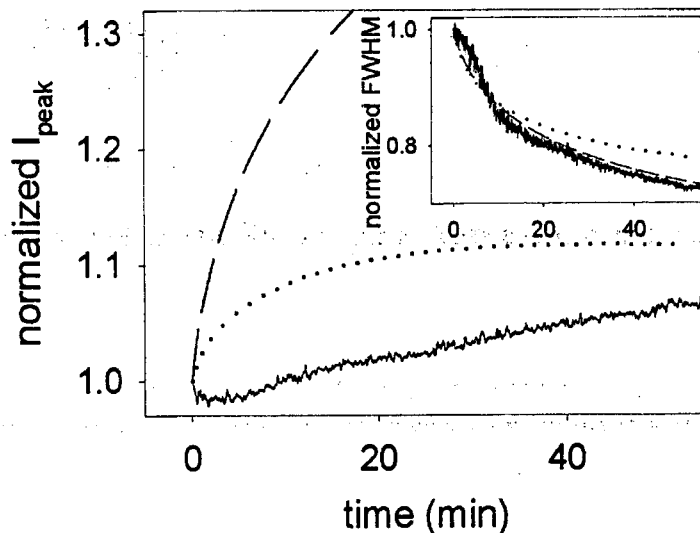


Fig. 6. Peak intensity at the sample output edge from experiment and simulations. The change in FWHM is shown in the inset. Dashed curves represents results without loss and dotted lines include a variable exposure-dependent loss of $1.6 \times 10^5 \Delta n$ dB/cm.

The shape of the beam emerging from the sample often changes slightly during the exposure, as in Fig. 1 where the beam grows a “shoulder” at late times in the experiment. Since the writing beam is Gaussian at the input face, this asymmetry must be due to some inhomogeneity in the sample. As the model used here assumes that the material is homogeneous, we cannot expect perfect agreement with the experimental data, especially at late times.

Clearly the model used in these simulations captures the essential physics of these self-writing experiments. Next, we use the experimental parameters deduced from this approach to achieve a deeper understanding of the self-written evolution which is occurring inside the material.

V. ANALYSIS

It has been shown previously [1] that in order to form a channel waveguide, the refractive index at the input face must grow large enough to counteract the diffraction of the beam, and focus the light in to an intensity maximum inside the material. This maximum is referred to as the *primary eye* and has so far always been viewed as a precursor to waveguide formation. Typically, this eye is located near the input face and so series expansions can be used to explore its behaviour at small distances in the propagation direction, z . This method has been applied to the planar geometry, for $p=1$ and 2, and in bulk for $p=1$ [2], corresponding to bulk photopolymer experiments from reference [7]. Here we extend this treatment to correspond to the two-photon absorption process in bulk cerium doped Ga-La-S glass, with the aim of achieving a greater understanding of our experimental results.

At early times the refractive index is too small to focus the incoming beam, and so a primary eye cannot form and the beam simply diffracts. We find from the series expansions that in our case the primary eye forms at time $T = T_0$ (as defined in equation (3)) that satisfies

$$\frac{\ln(4T_0)}{T_0} = \frac{2n_0}{k_0^2 a^2 \Delta n_s} \quad (5)$$

(for further details about this approach, see Ref [2]).

From equation (5) it can be seen that for any specific Δn_s , there exists a minimum writing beam width for an eye to form: if the beam is too narrow, the material cannot ever change index enough to counteract its diffraction. For the glass in our experiment, n_0 is approximately 2.5, and a good agreement with experiments occurs when Δn_s is taken to be 2.5×10^{-4} (see previous section). Using these values, equation (5) is plotted in Fig. 7 and the time at which the primary eye forms with four different choices of experimental parameters are marked A-D. Each of these cases is discussed below.

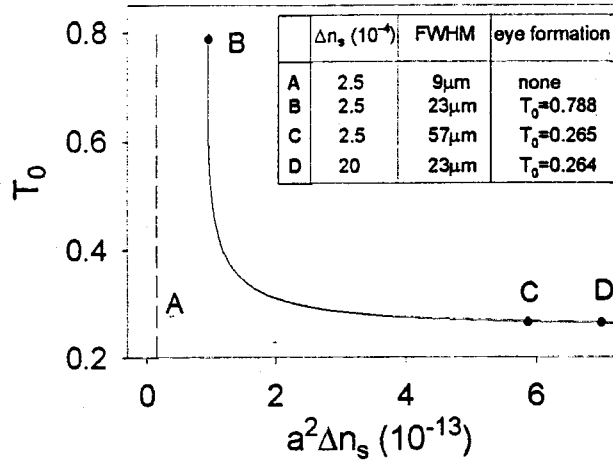


Fig. 7. The primary eye formation time (T_0) as a function of beam width and Δn_s . Dots show the formation time for four different choices of experimental parameters.

Case A: This case corresponds to the experiments in Fig. 2. It can be seen in Fig. 7 that the primary eye can never form using this beam width; and therefore a waveguide will not form. This is verified in Fig. 8 where the contour plot of the index distribution inside the material is plotted late in the evolution ($t = 195$ min in Fig. 5). Here the saturation value has been reached near the input face, no waveguide is formed and the index change only penetrates 1mm into the material.

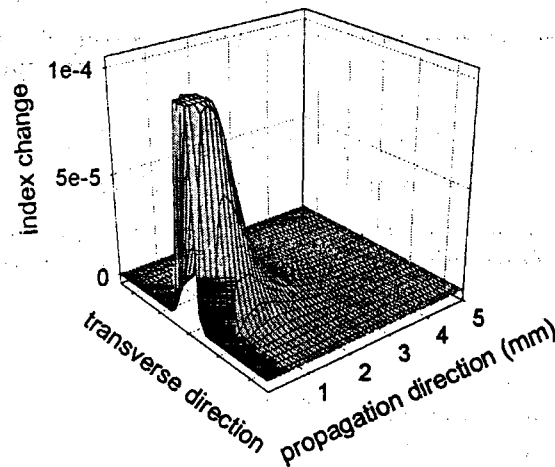


Fig. 8. Case A: The refractive index distribution within the material using a writing beam size of 9 μ m and $\Delta n_s = 2.5 \times 10^{-4}$. This corresponds to the simulation shown in Fig. 5 at $t = 195$ min.

Case B: Leaving the saturation index the same as in case A, but increasing the beam width to 23 μ m, equation (5) can be satisfied and an eye can form, however this will take a relatively long time ($T_0 = 0.788$ in Fig. 7). In Fig. 9 the refractive index distribution that evolves inside the material using these parameters is shown. It can be seen that a wider beam allows the structure to penetrate through the material.

Although a channel can form using this beam width, the primary eye has a very long formation time, and so the waveguide takes a long time to evolve, which is not ideal.

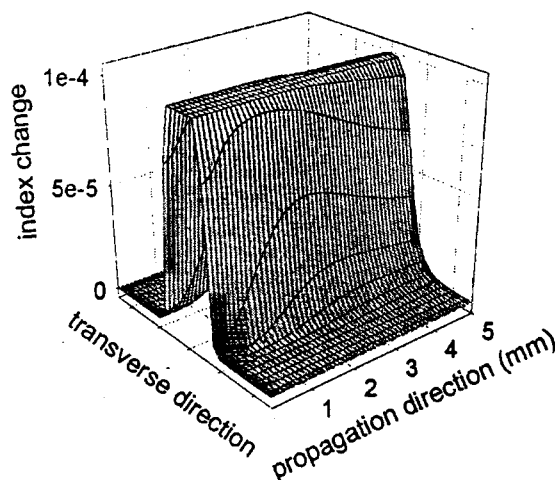


Fig. 9. Case B: The refractive index distribution when a beam of $23\mu\text{m}$ is used. Here a Δn_s of 2.5×10^{-4} is enough for a channel to form through the material, however this will require a very long time.

Case C: Here, the beam width has been increased even further to $57\mu\text{m}$. By doing this, the time at which the primary eye forms is reduced significantly to $T_0 = 0.265$. This would be the “ideal” experiment using cerium-doped Ga-La-S, where the saturation index of course is fixed. Recall in our simplified model that the same change in refractive index can occur regardless of the intensity. However experimentally, using such a wide beam, the power needs to be increased significantly in order to achieve sufficiently high refractive index changes (recall Fig. 4), and this has not yet been done experimentally.

Case D: Another way to make the primary eye form earlier in the evolution is to use a material with a larger saturation refractive index. For example, if $\Delta n_s = 2 \times 10^{-3}$ the formation time will be 0.264 using a beam width of $23\mu\text{m}$ (see Fig. 7). The index distribution that evolves inside the material for this case is shown in Fig. 10. It can be seen that a mature, uniform channel waveguide has formed through the material, and hence a large refractive index increase is favourable in self-writing. In this simulation, the FWHM decreased by a factor of 9, which is significantly larger than the change of 1.5 that was observed in the experiments shown in Fig. 2. This saturation index (2×10^{-3}) is achievable in other materials, and so this experiment should be practical in the future.

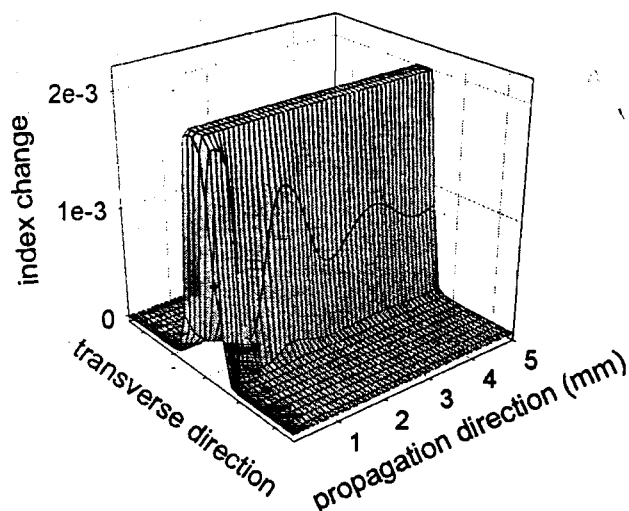


Fig. 10. Case D: The refractive index distribution when $\Delta n_s = 2 \times 10^{-3}$ and a writing beam of $23 \mu\text{m}$ is used, which results in the formation of a mature channel waveguide.

Information about the primary eye can be used to predict the parameters required to form a channel waveguide. It explains why a mature channel did not form in our experiments, where the narrow beams used prevent the primary eye from forming. In addition, it helps to determine which materials should be most suitable for future experiments.

V. DISCUSSION AND CONCLUSION

These experiments represent the first observations of self-written effects in a bulk glass. The refractive index within the cerium-doped Ga-La-S glass increased under illumination and so the diffraction of the propagating beam was decreased. By increasing the beam intensity, a large change in index occurred and the process proceeded faster. Numerical simulations of the process show excellent agreement with the experiments, which validates the model and indicates that we understand the basic principles of the process. Using this modelling we have shown that self-written channel waveguides can form in these bulk materials using appropriate parameters and conditions.

To obtain a deeper physical insight into our experimental results, analytical series expansions have been used. The behaviour of the primary eye was explored, and hence the formation of waveguides using different experimental parameters were analysed. It was found that using narrow beams ($< \sim 20 \mu\text{m}$), as in the experiments with cerium-doped Ga-La-S glass, the refractive index change in the sample, $\Delta n_s = 2.5 \times 10^{-4}$, was not large enough to form an eye and so create a waveguide through the material. Despite this limitation, the diffraction of the beam propagating through the material decreased significantly during the experiment and a weak taper was formed.

By introducing different dopants the photosensitivity of these glasses can be improved, which would enable bigger refractive index changes and therefore more sophisticated structures. Since these self-written structures are typically long-lasting,

waveguides in these glasses could be subsequently used to guide light at other wavelengths. Such compound glass materials transmit light in the far IR from 0.5 to 10 μm [17], and so long wavelength devices could be formed using this approach.

Although in this early work only Gaussian writing beams are considered, theoretical work shows that the shape of the resulting waveguide can be tailored via the writing beam [2]. This three-dimensional bulk geometry should enable great flexibility for creating complicated waveguide structures.

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