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Experimental and theoretical study of stable negative index gratings formed at 193 nm

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## **ABSTRACT**

We have demonstrated fast formation (~1500 pulses at ~1 J/cm²/pulse) of fibre gratings with high negative index modulations (~-3×10<sup>-4</sup>). These gratings were found to be far more stable than the gratings with positive index modulations formed at the early stage of the grating growth. We have also found that the maximum negative index modulations achieved do not depend on the pulse intensities, although the inverse of the time taken to reach the negative index modulation maximum varies linearly with the pulse intensities. This prompts us to use a three energy level system to model the photosensitivity in the boron-doped germanosilicate fibre. All the necessary parameters of the model can be determined from a single growth measurement of the average index change and the model's prediction fits well the measured index modulation growth. A complex grating decay process is also observed at elevated temperatures as predicted by the three energy level model. The thermal stability of both positive and negative index gratings in a Boron-co-doped germanosilicate fibre is characterised at fixed temperatures, so that the stability of such grating can be accessed for any writing fluence.

**KEYWORDS:** optical fibre photosensitivity, optical fibre gratings

# 1. BACKGROUND

The numerous applications of UV-written fibre Bragg gratings have generated a tremendous amount of interest in this area in recent years. Despite the large number of demonstrated devices based on such gratings, there has not been an established phenomenological model which a grating fabricator can use to predict grating growth and other characteristics. This has somewhat hampered the growth of the field. A better understanding of the process is urgently needed both for the optimisation of the fibre design and also for the optimisation of the grating manufacturing process.

In this paper, we have systematically studied grating growth in a boron-co-doped germanosilicate fibre using a ArF excimer laser at 193 nm and observed negative index modulations as high as  $\sim -3 \times 10^{-4}$ . We have also found that the maximum negative index modulations do not depend on the writing pulse intensities and the inverse of the time taken to reach the negative index modulation maximum is proportional to the pulse intensity. We propose a three energy level model for the process to account for our experimental observations. All the parameters of the model can be derived from a single average index change growth measurement. The model's prediction of index modulation growth fits well with the measured growth by adjusting only the writing fringe contrast. We have also confirmed that negative index grating is much more stable than the positive index gratings formed at the early stage of the grating growth [1]. This is attributed to a higher decay energy barrier for the level giving the negative index change.

Boron-co-doped germanosilicate fibres were found by Williams et al to be very photosensitive [2]. We have reported that saturated index changes of ~ 4 times larger than that in pure germanosilicate fibres can be obtained in these types of fibres [3] and have used these fibres regularly in our laboratories. In an earlier study when writing at 193 nm, we have found that a grating with positive index modulation was formed first and was then followed by a strong negative grating [4]. A similar type of grating growth has been observed previously when writing at 248 nm by Xie et al [5]. They have also found that the negative index grating is much more stable than the positive index grating formed at the early stage of the grating growth [1]. In our earlier study [3], we have observed that grating growth when writing at 193 nm is much faster than writing at 248 nm. This allows us to observe the saturation of index changes at various conditions within a realistic time scale for this study.

## 2. STUDY OF GRATING FORMATION AT 193 NM

Fig.1 gives the growth of the index modulation and corresponding average index change when writing with a pulse intensity of  $0.31 \text{ J/cm}^2$ . The horizontal axis is shown on a log scale to show the fast growth at the beginning. A pulse repetition rate of 20 Hz and a grating length of 1 mm were used for all the measurements in this study unless stated otherwise. A grating phase mask supplied from QPS Technology Inc was used. The Bragg wavelengths of all the gratings in this study were around 1530 nm. The exposure in fig.1 was long enough to cover all the features in the growth. The grating peak reflectivity and Bragg wavelength were monitored during writing to obtain both index modulation and average index change. The index modulation reached a positive maximum after  $\sim 0.5$  minute with a strength of  $\sim 1.0 \times 10^{-4}$  followed by a negative maximum at  $\sim 7$  minutes with a strength  $\sim -2.5 \times 10^{-4}$ . The index modulation decreased slowly toward zero after the negative maximum. The average index change reached its positive maximum after  $\sim 1$  minute and a saturation was reached after  $\sim 40$  minutes. The lines are theoretical fits obtained from our model which will be discussed later in this paper. It is clearly shown by the growth of the average index change in this figure that a fast positive index change was formed first and then followed by a slower negative index change.

To study the process in more details, we wrote a number of gratings at different intensities. These are plotted in fig.2. In this case, we stopped the writing process as soon as the negative index modulation reached its maximum. The vertical axis represents the total reflection of the gratings using a broad band LED. The final total reflection was normalised to the reflectivity of the grating at the end of the writing process. It can be clearly seen that the grating growth at high writing pulse intensities is much faster, albeit the saturated reflectivities do not vary very much with the writing pulse intensities. The process at some intensities was repeated several times to show the repeatability of the measurements. We plotted the inverse of the time taken to reach the negative index modulation maximum ( $T_{max}$ ) and the maximum negative index modulations against pulse intensities in fig.3. The growth rate of the negative index modulation ( $1/T_{max}$ ) was found to vary linearly with the pulse intensity. The index modulations of the corresponding gratings, however, do not depend significantly on the pulse intensity. This demonstrates that the grating growth is a linear process, depending only on the total fluence of the exposure.

## 2.2 Modelling of grating formation

These observations lead us to propose a three level energy system for the process where a positive index change is obtained when a lower level (level 2) is populated by depleting a ground level (level 1) and a negative index change is achieved when a higher level (level 3) is populated by depleting level 2 (see fig.4). A more realistic energy system would include many other intermediate levels with much shorter life times (<< 1s). We ignore these levels in this study for simplicity as we are only interested in levels with much longer life time (>1 s). No decay from levels 2 and 3 was observed during the time scale of the grating writing. We have, therefore, assumed that  $A_{21}$ ,  $A_{32}$  and  $A_{31}$  are zero. The model predicts a double exponential growth for the total index change

$$\Delta n_{tot}(t) = S_3 N \left( 1 + \frac{S_2 - S_3}{S_3} \frac{B_{12}}{B_{12} - B_{23}} e^{-B_{23} \rho t} + \frac{S_3 B_{23} - S_2 B_{12}}{S_3 (B_{12} - B_{23})} \right) e^{-B_{12} \rho t}$$
 {I}

where  $S_2$ ,  $S_3$ , N and  $\rho$  are index changes per unit population for levels 2 and 3, initial population at the ground level and writing beam intensity respectively (see fig.4 for the definitions of the other parameters).  $S_3$  is negative to account for the negative index change obtained by populating level 3.  $B_{23}$  is typically much smaller than  $B_{12}$ .  $\Delta n_{tot}(t)$  grows to a positive maximum at a rate of  $B_{12}\rho$  and then reaches a negative saturation at a rate of  $B_{23}\rho$ . The strength of the saturated negative index change is  $S_3N$ , independent of the intensity  $\rho$ . The time to reach the maximum positive average index can be simply derived from the equation 1.

$$T_{\text{max}} = \frac{1}{(B_{12} - B_{23}) \rho} \ln \frac{S_3 B_{23} - S_2 B_{12}}{(S_3 - S_2) B_{23}}$$
 [II]

The model predicts the observed linear dependence of growth rate on intensity and the constant saturated negative index changes at different pulse intensities. A fit to the average index growth in fig.1 was obtained with the following parameters:  $B_{12}\rho = 6.96 \text{ s}^{-1}$ ,  $B_{23}\rho = 151.2 \text{ s}^{-1}$ ,  $S_2N = 7.4$  and  $S_3N = -5.7$ . The best fit to the average index modulation was shown in fig.1 together with the measured growth. By assuming a writing fringe contrast

of 45%, we were able to predict the index modulation growth from the obtained average index growth. If the writing fringe is expressed as  $\rho = \rho_0(1 + m \cos(2\pi z/\Lambda))$ , where  $\rho_0$ ,  $\Lambda$ , m and z are average writing beam intensity, grating period, fringe contrast and length along the axis of the fibre,  $\Delta n_{tot}$  in equation 1 becomes both a function of z and t. The index modulation can be derived by simply taking the Fourier transform of  $\Delta n_{tot}(z,t)$ .

$$\Delta n_{\text{mod}}(t) = \frac{2}{\Lambda} \int_{\frac{\Lambda}{2}}^{\frac{\Lambda}{2}} \Delta n_{\text{tot}}(z,t) Cos(\frac{2\pi z}{\Lambda}) dz$$
 {III}

The fit is also shown in fig.1. The predicted index modulation growth fits well the measured data. The position of the negative index modulation peak was found to be sensitive to the fringe contrast in a more detailed study of the model. The error around the negative index modulation peak was possibly from a small change in contrast during writing. The index modulation will decay to zero after a long period of exposure due to the loss of index modulation contrast in the fibre. The maximum negative index modulation is predicted by the model to increase linearly with the writing fringe contrast up to a contrast of 95%.. A negative index modulation of ~-5.2×10<sup>-4</sup> should be achieved with a writing fringe contrast of 95%.

#### 2.3 Grating amplification by a subsequent uniform exposure

Amplification of a weak grating by a subsequent uniform exposure has been observed when writing with a 193 nm excimer laser by Dyer et al [6]. A damage mechanism was proposed to explain this. We have also studied this phenomenon. Several gratings were written with a different number of pulses (1 to 50) at 0.31 J/cm²/pulse. One of the measurements is shown in fig.5. The grating was written with 5 pulses. The grating was seen to decay first and then grew, followed by a second decay. The average index growth is also plotted in fig.5. It is clear that the effect is related to the change of growth rate in the average index (second order differentiation of the average index growth). When the growth rate increases, the index at the peak of the writing fringe grows faster than that at the trough. This leads to a growth of the index modulation. The horizontal axis in fig.5 is plotted on a linear scale to show the change in growth rate of the average index. A decrease in the growth rate will lead to a reduction of the index modulation. We found that the grating vanishes after the initial decay and does not survive to the amplification stage when it was written with a very small number of pulses (< 5).

## 2.3 Conclusions

To conclude this part of the study, we have systematically studied the index changes induced by a 193 nm excimer laser and modelled the process. The model fits well with all our experimental observations. Since similar grating growth was observed when writing at 248 nm albeit at a slower rate [1], a similar model may be used with relevant growth rates adjusted. The determination of all the necessary parameters requires only a single measurement of the average index growth, which can be performed easily without much attention paid to stabilising the grating writing apparatus. A further study is required to understand the microscopic details of the process.

## 3. STUDY OF GRATING DECAY AT ELEVATED TEMPERATURES

#### 3.1 Introduction

In the three level model, a positive index change is produced when a second level (level 2) is populated by depleting a ground level (level 1). A negative index is then created by populating a third level (level 3) from level 2. Populations at level 2 and level 3 will eventually diffuse into nearby trapping sites. The population at level 3 is able to access more stable traps than that of level 2, as it have been observed by Niay [1] in their study of these gratings formed at ~240 nm. Normally a grating will be formed with both levels 2 and 3 partially populated. A complex decay process is expected due to the difference in thermal stability of the two components.

## 3.2 Complex decay of gratings

When a grating is formed with predominantly level 2 populated, a simple decay is expected (see grating A in fig.6, a grating length of 1 mm is used to ensure uniformity of the gratings). As the population in level 3 increases (see grating B), the grating will decay to zero when the level 2 population decreases just enough to

balance the negative index change, this happening at 300°C for grating B. Beyond this point, the grating is formed by an overall negative index change and is expected to increase when the level 2 population decreases further (see grating B at 400 °C). This effect is much more obvious for grating C when such a balance between the positive and negative index changes is achieved at the beginning of the decay process (there is almost no grating in the fibre when written). For the overall negative index grating (D and E), the decrease in level 2 population at lower temperature, will cause the grating to grow until the level 3 population starts to decrease. Note in grating F when the population is mainly in level 3, hardly any grating decay was observed until 300 °C.

## 3.3 Thermal decay at fixed temperatures

For a grating with both level 2 and 3 populated, a pre-annealing procedure can be done to remove the population at level 2 to achieve a more stable grating. It is therefore important to study the decay of levels 2 and 3 in more detail. Thermal decay for level 2 was performed with gratings exposed for ~ 30 seconds to ensure that level 3 is not significantly populated. As for weak gratings ( KL<0.5), total reflection from a grating is proportional to ( κL)<sup>2</sup>. For the experiment in fig.7, the gratings were (18±3)% reflection. Total reflection is monitored and then converted into normalised  $\kappa$ . The results are shown in fig.7. A power law function  $1/(1+At^{\alpha})$  was fitted to each curve to obtain the parameter A and (see [7] for more details). A and  $\alpha$  are found to be 0.00173e<sup>0.00824T</sup> and 0.0153e<sup>0.00575T</sup> respectively [see fig.8]. A deviates substantially from a linear relation to T as proposed by Erdogan et al [7]. The positive grating is expected to decay by 11% over 25 years. A pre-annealing at 300 °C for 10 minutes will remove 40% of the population in level 2 (the less stable part of the population in level 2) without having too much effect on the level 3 population. An accurate stability measurement of the level 3 gratings is much more difficult to obtain, as there is always some non-zero population in level 2. Even worse is the fact that at the trough of the writing fringe, the fibre is exposed to less fluence than that at the peak. Therefore, the proportion of level 2 population varies from one part of the grating to another. A thermal stability test for gratings written with ~30 minutes of exposure is shown in fig.9. The gratings were 0.5 mm long and (17±3)% reflection. The decay at the beginning of each curve shows some abnormal decay, perhaps affected by the decay of the small amount of level 2 population. The stable nature of the negative index change is obvious (see the curve at 400°C). In any case, a pre-annealing stage can be performed to remove the unstable part of the positive index change to achieve much more stable gratings.

### 3.4 Conclusions

We have observed the complex decay behaviours predicted by the there energy level system. The stability of both positive and negative index gratings have been characterised. The much stable negative index gratings will potentially find many sensor applications and applications where high temperature stability is required.

## 4. ACKNOWLEDGEMENTS

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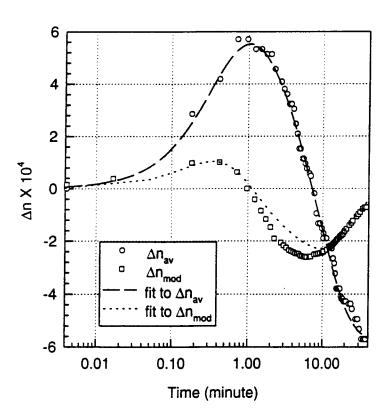


Figure 1 The growth of index modulation and average index when writing with a KrF excimer laser at 0.31 J/cm<sup>2</sup>/pulse together with the fits from the proposed model.

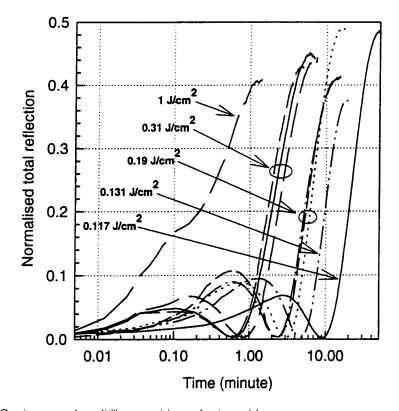


Figure 2 Grating growth at different writing pulse intensities.

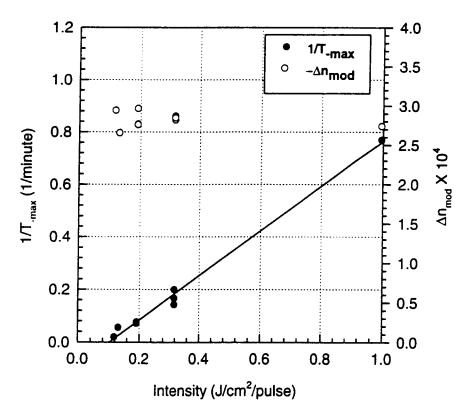


Figure 3 The dependence of the growth rate and maximum index modulation of the negative index gratings on different writing pulse intensities.

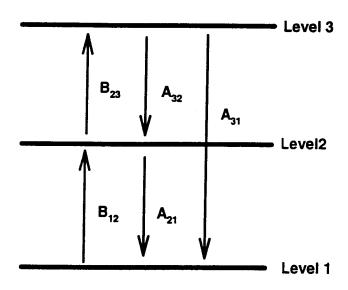
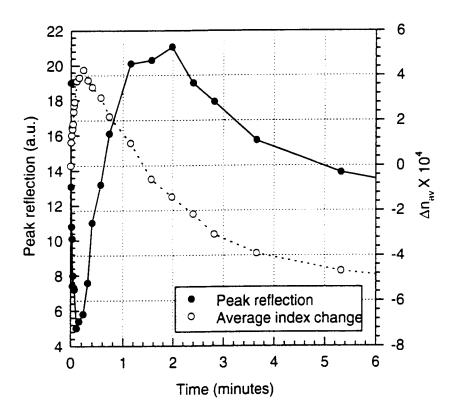


Figure 4 The energy diagram of the proposed model.



**Figure 5** The amplification of a grating by a uniform exposure at 0.31 J/cm<sup>2</sup>/pulse. the grating was written by five 193 nm pulses at 0.31 J/cm<sup>2</sup>/pulse.

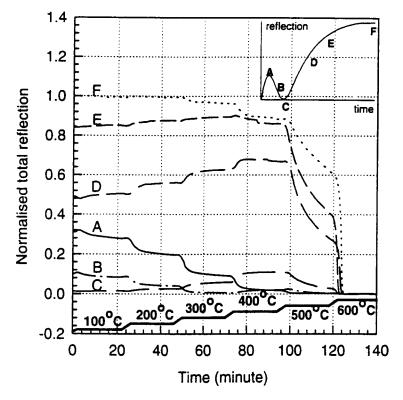


Figure 1 Complex decay behaviour of gratings.

Figure 6 Complex decay behaviour of gratings.

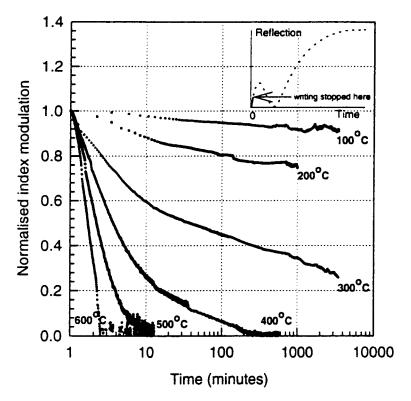


Figure 7 Decay of positive index gratings.

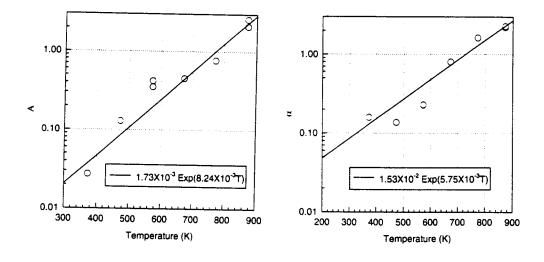


Figure 8 Dependence of A and  $\alpha$  on the inverse of the temperatures.

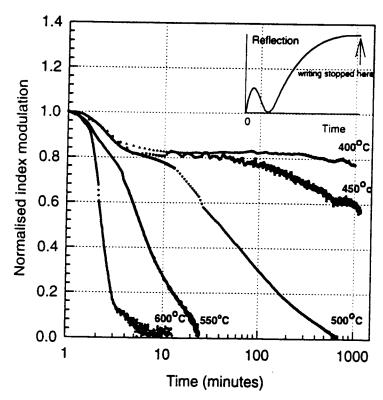


Figure 9 Decay of negative index gratings.