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Stress-dependent growth kinetics of ultraviolet-induced refractive index change and defect centers in highly Ge-doped SiO_2 core fibers

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The evolution of the index change of type-IIa gratings observed in 28 mol % Ge–SiO_2 core fibers with 1.8 μm core diameter under various strains was measured from the optical spectra, and the induced defects at high and low strains were studied with electron spin resonance. Data will be presented to show that the index modulation (Δn_{mod}) of type-IIa gratings is likely associated with Ge E' centers. © *1999 American Institute of Physics.* [S0003-6951(99)04141-8]

Fiber Bragg gratings (FBGs) are emerging as critical components for telecommunication and sensor applications.[1] So far, three types of fiber Bragg gratings have been reported.[1] Type I gratings exhibit a monotonic increase of both the mean effective refractive index and the induced index modulation under ultraviolet (UV) exposure.[1,2] Although their spectral characteristics are excellent, their thermal stability is limited to 300 °C. Type II FBGs, formed with a single excimer pulse, feature large UV induced refractive index changes (typically 10^{-2}) and high thermal stability to 800 °C.[3] Nevertheless the poor spectral characteristics of type II FBGs fabricated to date prevent them for technological uses. On the other hand, type-IIa gratings display similar spectral properties as type I FBGs, but are thermally stable to 550 °C.[2] Thus, type-IIa gratings may be suitable for high temperature applications, and understanding the mechanism responsible for type-IIa FBG formation is important for their technological usage.

Typically, the most relevant feature of type-IIa gratings is the complex behavior of Δn_{mod} and $\langle \Delta n_{\mathrm{eff}} \rangle$ with increasing UV exposure time. Indeed, a partial or total bleaching of the saturated type I grating is followed by the formation of a new type-IIa grating, suggesting an inverse relationship between the index modulation of these two types of gratings. So far, type-IIa FBGs have been reported in highly Ge-doped, small core silica fibers, and more recently in boron-codoped germanosilicate and nitrogen-doped fibers.[2,4,5] Based on the experimental results, a model was proposed to explain the complicated grating growth: Briefly, the bleaching rate of the oxygen deficient center (ODC), which is responsible for the formation of type I FBGs, is presumed to be larger than that of unknown centers that produce the negative index change responsible for the formation rate of the type-IIa grating spectrum. Although the model explains the observed phenomena, it does not elucidate the microscopic origin of the type-IIa grating precursors.

It has been reported[6] that the thermal stability of type-IIa gratings is similar to that of the paramagnetic Ge E' center, implying that the Ge E' center is associated with type-IIa gratings. Also it has been shown recently that the complex dynamics observed in fibers exhibiting type-IIa gratings were strongly dependent on the strain applied to the fiber at the time of the photoimprinting.[7] Studies of the growth kinetics of paramagnetic defects under similar applied strains as those of type-IIa gratings can then provide additional information regarding the contribution of paramagnetic defects to type-IIa gratings.

The aim of this letter is to report further investigations undertaken to determine if a relationship exists between the formation of Ge E' centers and the appearance of type-IIa grating spectra and their respective dependencies on strain applied during FBGs inscription. Furthermore, it has been recently shown in short period gratings[8] that the Ge(1,2) centers contribute only to an increase of average UV-induced refractive index change, $\langle \Delta n \rangle$. We will also report how the growth kinetics of the Ge(1,2) centers are affected by external strain and the influence of strain on $\langle \Delta n \rangle$.

Single mode fibers with a core diameter of 1.8 μm doped with 28 mol % Ge were used in this study, and the strain applied to fibers during UV exposure was the parameter of the experiment. The fibers were exposed to a KrF excimer laser through a phase mask (Lasiris Corp. Canada) for FBG fabrication. Grating reflectivity and Bragg wavelength shift were monitored in real time using a broad band Er-doped superluminescent source and an optical spectrum analyzer (AQ6315A, ANDO, Japan). Photoinduced paramagnetic defects were studied by X-band (≈9.4 GHz) electron spin resonance (ESR) at room temperature using a Bruker ER 200D-SRC spectrometer in second harmonic mode operation. The ESR samples consisted of 2-cm-long pieces of fiber of total length 60 cm that were uniformly exposed with the same power used to write the FBGs at high strain ($\Delta L/L = 9.84\times 10^{-3}$) and low strain ($\Delta L/L = 3.85 \times 10^{-4}$).

Figure 1 shows the evolution of the UV induced paramagnetic defects for high and low strains. One can see that the concentration of Ge E' increases faster at high than at low strain. The solid lines in Fig. 1 are the fit of $[\mathrm{Ge}\,E']$ to a saturating exponential

$$[\mathrm{Ge}\,E'] = b_{\mathrm{Ge}\,E'}[1-\exp(-k_{\mathrm{Ge}\,E'}P)], \tag{1}$$

where P is the number of pulses, $b_{\mathrm{Ge}\,E'}$ and $k_{\mathrm{Ge}\,E'}$ are the parameters of the saturating exponential equal to, respectively, $3.5\times 10^{19}/\mathrm{cm}^3$, and 0.000 645 at high strain and $3.0 \times 10^{19}/\mathrm{cm}^3$ and 0.000 172 at low strain. Furthermore, one

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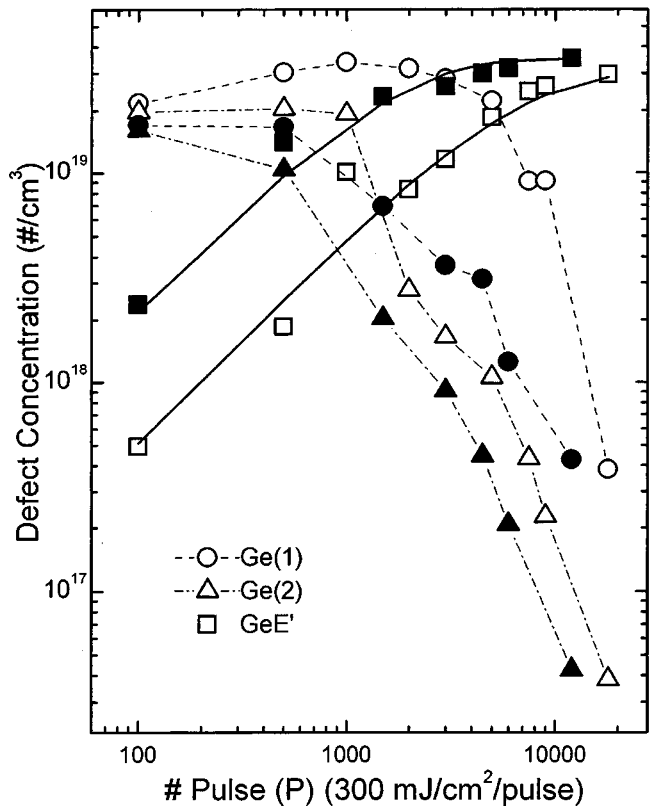

FIG. 1. Paramagnetic defect center concentration measured as a function of the number of laser pulses at high strain$=9.84\times10^{-3}$ (solid points) and low strain$=3.85\times10^{-4}$ (open points). Solid lines are fits to Eq. (1).

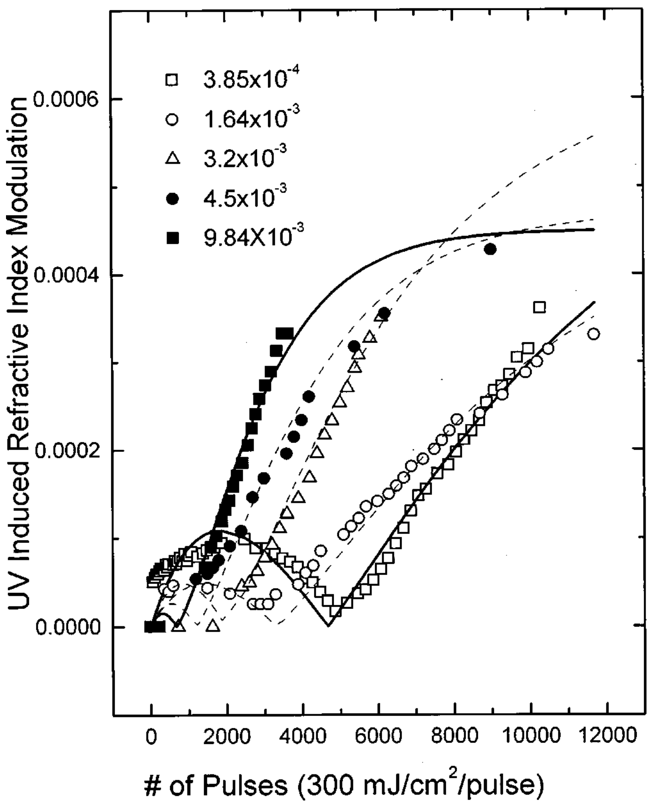

FIG. 2. Index modulation of FBGs at various strains as a function of the number of laser pulses. Solid lines are fits to Eq. (2) using the growth kinetics of the Ge E' center (Fig. 1) for the type-IIa grating and negative saturated exponential growth kinetics for the tension increase and densification.

can see that at low strain, the Ge(1) and Ge(2) concentrations increase up to 1000 pulses and then decrease, while at high strain the decrease in their concentrations is observed when P is greater than 100.

The UV-induced Δn_{mod} is shown as a function of P under various strains in Fig. 2. Clearly observed at low strain is the fact that growth of type-IIa gratings follows a total bleaching of type I gratings. However, at high strain, as previously observed in Ref. 7, type I gratings are hardly observed. The growth kinetics of Δn_{mod} observed at high and low strains in Fig. 2 can be fit with the following equation:

$$\Delta n_{\mathrm{mod}}=|\beta[\mathrm{Ge}\ \ E']+\Delta n_{\mathrm{oth}}[1-\exp(-k_{\mathrm{oth}}P)]|, \tag{2}$$

where $\beta(=6\times10^{-23})^{9}$ is the contribution, per unit concentration, of Ge E' (measured by ESR spectroscopy) to Δn_{mod}, and Δn_{oth} stands for the total contribution to the index modulation associated with the UV-induced tension increase[10] and densification.[11] The solid lines in Fig. 2 are the fits using Eq. (2) with Δn_{oth} and k_{oth} equal, respectively, to -1.65×10^{-3} and 8.85×10^{-4} for high strain and -1.21×10^{-4} and 3.7×10^{-4} for low strain. The dotted lines in Fig. 2 are also fits of the growth kinetics of Δn_{mod} at intermediate strains using similar saturated exponential growth of Eq. (2). Since the data of Fig. 2 show that the maximum of index modulation of type-IIa gratings is greater than that of type-I gratings, the type-IIa grating is to be associated with the larger in absolute value of the two terms in the fits using Eq. (2). Since $|\beta[\mathrm{Ge}\,E']|$ is larger than $|\Delta n_{\mathrm{oth}}[1-\exp(-k_{\mathrm{oth}}P)]|$, we then associate the Δn_{mod} of type-IIa gratings with Ge E' centers. The good correlation between the increase in $[\mathrm{Ge}\,E']$ with applied strain and the early appearance of the type-IIa grating spectrum shows also that the first term in Eq. (2) is associated with the index modulation responsible for the type-IIa grating. In addition, this assignment is in agreement with the report of similar thermal stability of the Ge E' center[6] and Δn_{mod} of type-IIa gratings.[7] It is to be noted in passing that the growth kinetics of Eq. (2) are mathematically identical to that proposed in Ref. 2.

Both positive and negative contributions to Δn_{mod} are necessary to account for the complicated growth in fibers that exhibit type-IIa gratings because their growth always follows a partial or total bleaching of saturated type-I gratings. However, in fitting the growth kinetics using Eq. (2), no absolute assignment of the sign of each term can be made as long as their relative contributions are the inverse of each other. In this sense, it is possible that the index change of type-IIa gratings associated with Ge E' centers is negative due to unknown phenomena not yet elucidated, although it is anticipated that Ge E' centers contribute a positive Δn_{mod} through the Kramers–Kronig relationship due to their optical absorption band in the UV region. However, the data of Fig. 3 obtained at $\Delta L/L=4.5\times10^{-3}$ in which only type-IIa gratings are observed show that the associated Bragg wavelength shift during its growth is positive. This implies that the contribution of Ge E' to the Δn_{mod} of type-IIa gratings is likely positive. [The decrease in Bragg wavelength that occurred at the saturation of type-IIa gratings seems correlated with the drastic decrease in population of Ge(1) and Ge(2) centers, as Fig. 3 illustrates, in agreement with the report in Ref. 8.] The Δn_{oth} observed in our fibers is then negative. This conclusion

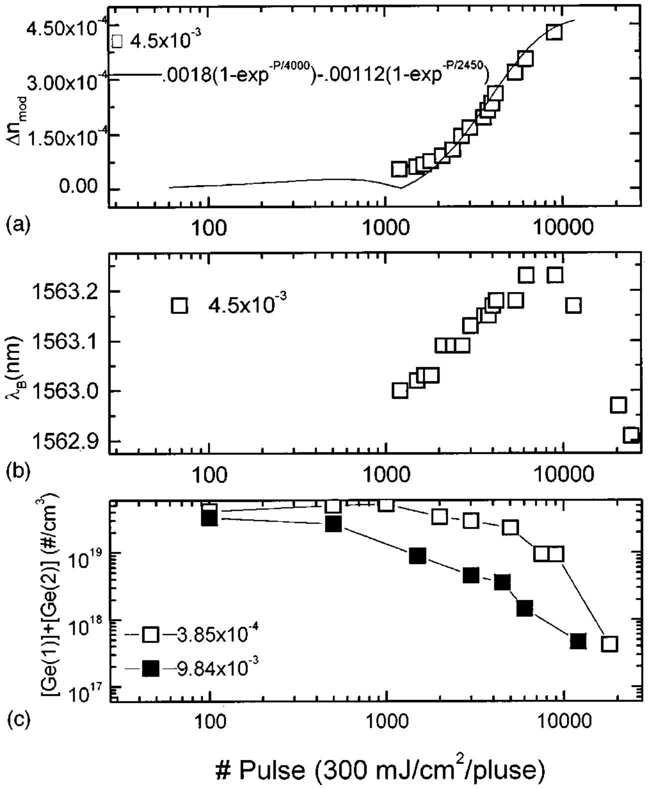

FIG. 3. Change in the (a) index modulation, Δn_{mod}, (b) Bragg wavelength, λ_B, of FBGs at strain $\Delta L/L = 4.5\times10^{-3}$, and (c) the sum of the concentrations of Ge(1) and Ge(2), as a function of number of KrF laser pulses.

can be explained by assuming that the negative index modulation associated with UV-induced tension increase is greater than the positive index modulation associated with densification in fibers that exhibit type-IIa gratings. In standard single mode photosensitive fibers with ≈8 μm core diameter, the negative Δn_{mod} associated with UV-induced tension increase is smaller than the positive Δn_{mod} associated with UV-induced densification, resulting in an overall positive Δn_{oth} and the observed monotonic growth in index modulation. The report that type-IIa photosensitivity is not observed in preforms[12] is also consistent with this result since the UV-induced tension increase in a preform is anticipated to be much less than the densification. In passing, we note that the positive Bragg wavelength shift observed here during the growth of type I gratings at low tension [correlated with the growth of Ge(1,2) at low fluence, see Fig. 1] and the negative Bragg wavelength shift during the growth of type-IIa gratings [correlated with the bleaching of Ge(1,2) at high fluence, see Figs. 1 and 3] can be attributed to the contributions of Ge(1) and Ge(2) to the average index of FBGs, as previously reported.[8]

In summary, we have found that at high and low strains, the growth kinetics of type-IIa gratings in 28 mol % Ge-doped SiO_2 fibers with 1.8 μm core diameter can be fit using the observed growth kinetics of the Ge E' centers. This suggests an association of Ge E' centers with the index modulation of type-IIa gratings, in agreement with their reported similarity in thermal stability.[6] The report that type-IIa gratings are observed in Ge–SiO_2 fibers with small core diameters but not in preforms can also be understood. The positive Bragg wavelength shift during the growth of the negative Δn_{mod} of type-I gratings and the negative Bragg wavelength shift observed after the saturation of the positive Δn_{mod} of type-IIa gratings can be attributed to the growth and bleaching kinetics of Ge(1) and Ge(2) centers, that contributes to the uniform index of short period gratings.[8]

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