Photoinduced refractive-index changes in germanosilicate fibers

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Photoinduced guided index changes approaching 10^{-4} in the range 488–784 nm, measured using a simple interferometric technique, are reported in germanosilicate single-mode optical fibers exposed to the 488-nm line of an Ar^+ laser running multifrequency. The wavelength dependence and dynamics of the writing process are characterized, and the material dispersion of the induced $\Delta n(\lambda)$ is shown to be weak. The effect is placed in the context of related research on color centers in these fibers, and two different mechanisms are proposed that lead to quantitative estimates in rough agreement with the measured Δn values.

The holographic formation of Hill reflection gratings in certain germanosilicate fibers at blue–green wavelengths has been reported by several authors.[1,2] We have recently utilized the effect to realize a bandpass transmission filter in a Sagnac loop.[3] The published experimental data indicate that the maximum index modulation obtainable is less than 10^{-7}. Thus it was a surprise when Saifi *et al.*[4] recently reported index changes Δn as high as 5×10^{-5} in a directional coupler exposed to intense femtosecond switching pulses at 620 nm. This result prompted us to investigate the average Δn induced in germanosilicate fibers exposed to cw light at 488 nm. To this end we have developed a simple and sensitive experimental setup that permits characterization of the effects of different probing–writing wavelengths and intensities.

Two fused tapered couplers were fabricated using a germanosilicate single-mode fiber (cutoff at 450 nm, N.A. = 0.26, mode-spot area 1.814 μm^2 at 488 nm) that was known to be photosensitive. Both had splitting ratios of 95:5 at 488 nm and 83:17 at 633 nm, and they were spliced together in the experimental arrangement shown in Fig. 1. Coupler 1 combines prism-selected 488-nm light from an Ar^+ laser with 633-nm light from a He–Ne laser; this light is then split by coupler 2 and delivered to ports 2c and 2d, which are cleaved to have lengths identical to within a few millimeters. High-visibility interference is obtainable at port 2b, regardless of the coupler splitting ratio. Any relative change in core refractive index in the two fibers (which will occur if there is an intensity-dependent Δn) will cause a periodic cycling up and down of each wavelength at port 2b. In order to minimize thermal effects, the fibers leading from ports 2c and 2d were placed in a narrow channel in an aluminum enclosure.

Low-power (10-μW) blue light was launched in initially, giving an in-core intensity of 5.5 $\mu W/\mu m^2$ in fiber 2c. No change was seen in either of the reflected light signals detected at port 2b, even when monitoring over tens of minutes. On increasing the power of the blue light, however, both reflected signals were observed to cycle through bright and dark fringes, as shown in Fig. 2, indicating a changing Δn. On blocking the blue light, the reflected red signal jumped approximately 5% of a fringe to a new level but thereafter remained constant until the blue light was unblocked again, when it returned to its original level and recommenced cycling through the fringes. This jump corresponds to an index change of 3.3×10^{-8} at 633 nm, of the same order as the thermal-index change caused by core heating[5]; spontaneous changes in the color-center population may also play a role.[6]

The intensity was then increased in discrete steps, dwelling at each new level until the cycling had significantly slowed before proceeding to the next value. The initial rates of index change $d|\Delta n|/dt$ at each stage are plotted against the in-core intensity at 488 nm in Fig. 3. The rate at 633 nm is always ~73% of that at 488 nm. The index change was observed to saturate slowly with time (Fig. 4); it was several hours before a near-stationary point was reached. After 8.5 h the index change obtained at an in-core intensity of 60 $mW/\mu m^2$ was 7.27×10^{-5} at 488 nm. The experiment was repeated with fresh fiber and couplers (splitting ratios 95:5 at 488 nm and 70:30 at 784 nm), using a 784-nm laser diode as a probe in place of the He–Ne laser. The index change observed at this wavelength was 40% of the change at 488 nm.

A variety of effects [e.g., color-center-induced

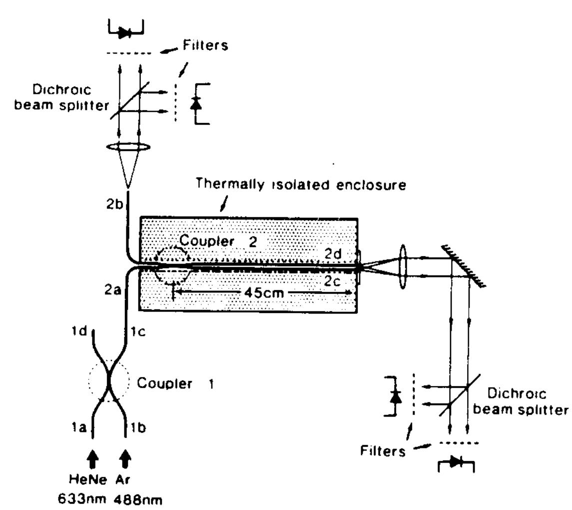

Fig. 1. Experimental arrangement.

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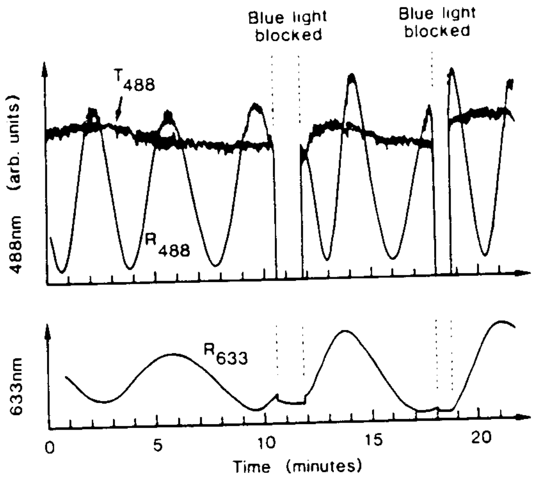

Fig. 2. Cycling of the fringes, including relaxation and recovery after turning the 488-nm laser light off and on. R_{633} and R_{488} are the reflected signals at port 2b.

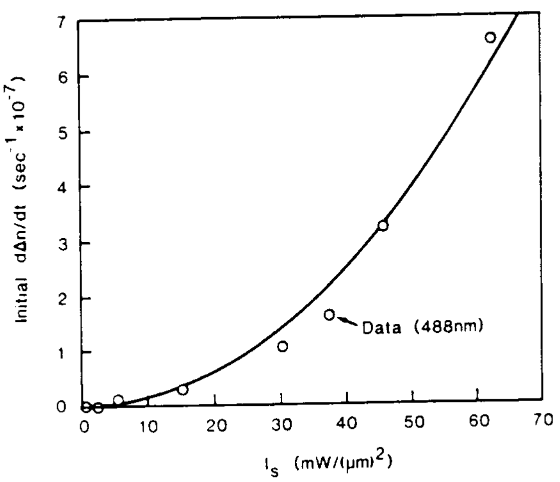

Fig. 3. Initial rate of increase of Δn versus the intensity level at 488 nm. The solid curve is a quadratic fit.

losses[6] and excitation-poled $\chi^{(2)}$ values[7]] in the blue-green spectral region are driven by two-photon absorption (TPA) near 480 nm to oxygen-deficient Ge–Si bond sites associated with an absorption band at 240 nm in the ultraviolet. Intensities of the order of only a few milliwatts per squared micrometers at 488 nm are sufficient to cause significant Ge–Si bond breakage, charge release, and retrapping at neighboring Ge sites to form additional absorption centers in the glass. This leads to changes in the fiber's absorption spectrum in the ultraviolet; the absorption band at 240 nm is partially bleached out by Ge–Si bond breakage, and the electrons released in this process are trapped at other defect sites, forming Ge(1) and Ge(2) color centers with absorption bands at 281 and 213 nm, respectively.[8]

It is reasonable to expect, assuming that the supply of Ge–Si bonds is far from exhausted, that the initial $d|\Delta n|/dt$ rate at each new intensity I_{488} should scale as I_{488}^2. This is because Ge–Si bond breakage is driven by TPA at 488 nm. The function $d|\Delta n|/dt$ (initial) = $K \times I_{488}^2$ yields a fair fit to the data for $K = 1.56\ \mu m^2/$ mW-sec (see Fig. 3), which supports the proposed mechanism.

The experimentally measured effective index changes were $\Delta n(633) = 0.73\Delta n(488)$ and $\Delta n(784) = 0.40\Delta n(488)$. Taking account of the fact that only a proportion of the guided light is present in the photosensitive core, the index change of the core material Δn_c is approximately $\Delta n_c(\lambda) = \Delta n(\lambda)/(1 - \exp\{-[0.83a/w(\lambda)]^2\})$, where $a = 0.618\ \mu m$ is the core radius and w is the HWHM Gaussian spot radius (a function of wavelength). For the measured LP_{11} mode cutoff of 450 nm, $w(488) = 0.76\ \mu m$, $w(633) = 1.00\ \mu m$, and $w(784) = 1.45\ \mu m$. This yields $\Delta n_c(488) = 1.8 \times 10^{-4}$, $\Delta n_c(488) = 1.8 \times 10^{-4}$, $\Delta n_c(633) = 2.0 \times 10^{-4}$, and $\Delta n_c(784) = 1.9 \times 10^{-4}$, with $\Delta n(488)$ being set at 7.3×10^{-5}. Owing to uncertainties in measuring the cutoff wavelength and nonuniformity in the intensity profile across the core, these values are accurate to perhaps $\pm 10\%$.

Our first mechanism is based on the idea that photoinduced changes in the absorption at 213, 240, and 281 nm give rise to the observed $\Delta n(\lambda)$ through the Kramers–Kronig principle. If we assume λ to be sufficiently far off resonance, the following three-term differential Sellmeier expression for $\Delta n(\lambda)$ may be used to fit the experimental data:

$$\Delta n(\lambda) = \sum_{i=213,\,240,\,281} A_i/[1 - (\lambda_i/\lambda)^2]. \tag{1}$$

If we assume Gaussian line shapes, the changes in peak absorption $\Delta\hat{\alpha}_i$ at each transition could then be related to the A_i values as

$$\Delta\hat{\alpha}_i = (E_i 2\pi^{3/2}/W_i\lambda_i)A_i, \tag{2}$$

where E_i is the transition center frequency in electron volts. For the fiber used, the maximum absorption induced at 488 nm by 100 mW/μm^2 of 488-nm light is of the order of 1000 dB/km. From Friebele and Griscom,[8] the FWHM absorption linewidths W at 213, 240, and 281 nm are $W_{213} = 0.9$ eV, $W_{281} = 1.97$ eV, and W_{240} (estimated from the absorption plot) = 0.22 eV.

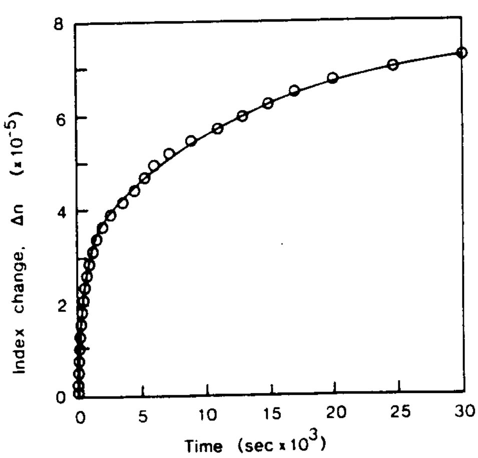

Fig. 4. Saturation of $\Delta n(488)$ with time at a constant intensity of 60 mW/μm^2. The fit (solid curve) was obtained using two decaying exponential rates: 1.87×10^{-3} and 7.46×10^{-5} sec^{-1}. Approximately the same values produced a good fit for $\Delta n(633)$.

The narrowness of the peaks at 213 and 240 nm means that the absorption induced at 488 nm will be due overwhelmingly to the creation of Ge(1) centers at 281 nm. This allows us to estimate that $\Delta\hat{\alpha}_{281} = 8.1\ \mathrm{m}^{-1}$, which from Eq. (2) gives $A_{281} = 9.2 \times 10^{-8}$. Adjusting $\Delta n_c(488)$, $\Delta n_c(633)$, and $\Delta n_c(784)$ to yield this value of A_{281} gives finally $A_{240} = -1.05 \times 10^{-4}$ and $A_{213} = 2.74 \times 10^{-4}$ for $\Delta n_c(488) = 2 \times 10^{-4}$, $\Delta n_c(633) = 1.864 \times 10^{-4}$, and $\Delta n_c(784) = 1.8 \times 10^{-4}$. These values of A_i lead from Eq. (2) to $\Delta\hat{\alpha}_{213} = 9.25 \times 10^4\ \mathrm{m}^{-1}$ and $\Delta\hat{\alpha}_{240} = -1.15 \times 10^5\ \mathrm{m}^{-1}$.

Using Smakula's approximation[9] for Gaussian linewidths,

$$\Delta N_i = [8.7 \times 10^{20} n/(n^2 + 2)^2] W_i \Delta\hat{\alpha}_i / f_i\ \mathrm{m}^{-3}, \qquad (3)$$

where $n = 1.458$ is the refractive index and f_i is the transition probability [$f_{281} = 0.42$ and $f_{213} = 0.77$ (Ref. 8); f_{240} is not given in Ref. 8], the population changes ΔN_i at 213 and 281 nm may be estimated as $\Delta N_{213} = 3.3 \times 10^{25}\ \mathrm{m}^{-3}$ and $\Delta N_{281} = 1.2 \times 10^{22}\ \mathrm{m}^{-3}$. Assuming that all the electrons released from broken Ge–Si bonds end up in Ge(1) and Ge(2) traps, $\Delta N_{240} = -(\Delta N_{213} + \Delta N_{281}) \approx -\Delta N_{213}$, which enables us to estimate the transition probability of the 240-nm transition (from Smakula's equation) as $f_{240} = 0.23$. Assuming further that roughly half the GeO_2 molecules are oxygen deficient, then the 15 mol. % of $GeO_{3/2}$ in the fiber core sets the upper limit of Ge centers at $3 \times 10^{27}\ \mathrm{m}^{-3}$. This indicates that $|\Delta N_{240}| \le 1.5 \times 10^{27}\ \mathrm{m}^{-3}$, which is in accordance with our estimate. It is interesting that this model predicts a much larger Ge(2) than Ge(1) population. This is plausible, since Ge(2) traps cannot be bleached by single-photon absorption at 488 nm and will prevent the electrons from reaching the Ge(1) traps.

The second mechanism that we propose involves the formation of permanent electric dipoles in the glass. The breakage of the Ge–Si bonds by TPA yields positively charged Si^+ sites (the Ge–E′ centers) and a free electron. The Si^+ ion is fixed in the glass matrix, whereas the electron has enough energy to escape and get trapped at neighboring Ge(1) or Ge(2) sites. Each resulting dipole will produce a static dc polarization field that extends many molecular spacings out. For example, at $r = 1$ nm away from the dipole center the field will be approximately $E_{\mathrm{dip}} = (qa/4\pi\epsilon_r\epsilon_0 r^3) = 145\ \mathrm{V}/\mu\mathrm{m}$ for a dipole spacing $a = 0.3$ nm. This frozen-in electric field will yield a local $\Delta n(\mathbf{r})$ that is proportional to $\chi^{(3)} E_{\mathrm{dip}}^2(\mathbf{r})$; the change in refractive index detected by a guided mode is then $\Delta n(\mathbf{r})$ averaged over the defect volume. Ignoring for heuristic reasons the tensorial nature of $\chi^{(3)}$, it is possible to estimate Δn as

$$\Delta n = [3\chi^{(3)}/2\sqrt{n_0}] \iiint E_{\mathrm{dip}}^2(\mathbf{r}) \mathrm{d}V \Big/ \iiint \mathrm{d}V, \qquad (4)$$

where the integrals are over the average volume around a dipole. The trick now is to note that the integral of $\epsilon_r\epsilon_0 E_{\mathrm{dip}}^2/2$ is the total energy stored in the dipole field, which cannot be greater than $2h\nu$ per excited carrier, the quantum of energy absorbed by TPA. This allows us to rewrite Δn as

$$\Delta n \le [3\chi^{(3)} N_{\mathrm{od}} h\nu / \epsilon_0 \epsilon_r \sqrt{n_0}], \qquad (5)$$

where N_{od} is the number density of oxygen-deficient Ge–Si sites and hence the upper limit of potential dipoles per unit volume; $1/N_{\mathrm{od}}$ is the average empty volume surrounding a single dipole. With $\Delta n_c = 1.8 \times 10^{-4}$, $\epsilon_r = 3.7$, $n_0 = 1.458$, and $\chi^{(3)} = 10^{-22}\ \mathrm{m}^2/\mathrm{V}^2$ and assuming that perhaps half of $2h\nu$ is stored in the dipole field, the number of dipoles needed per unit volume turns out to be $N_{\mathrm{od}} = 1.2 \times 10^{26}\ \mathrm{m}^{-3}$. As shown above, the Ge number density is approximately $3 \times 10^{27}\ \mathrm{m}^{-3}$. In order to yield the observed Δn_c, only 4% of these need be both oxygen deficient and activated to yield a dipole.

Of these two explanations, we favor the Kramers–Kronig mechanism since (i) the population change ΔN_{240} needed is smaller and (ii) it is likely that a much smaller fraction of $2h\nu$ than 50% will end up in E_{dip}.

It is interesting to note that the Δn reported by Saifi *et al.*[4] at 620 nm may be explained if the Ge–Si bonds, which exhibit a weak subsidiary absorption at 320 nm, are broken by TPA at 620 nm.

Attempts were made to induce Δn values using mode-locked Q-switched Nd:YAG pulses at 1.064 μm. No change in index was detected, which is encouraging considering that nonphotosensitive reflection filters are needed at these wavelengths. The low material dispersion of Δn_c suggests that substantial index changes [Eq. (1) predicts $\sim 1.9 \times 10^{-4}$] may be achievable beyond 1 μm; this offers the tantalizing prospect, using side writing, of $(\lambda/2n_0\Delta n) =$ 3-mm-long 100% reflection filters at 1.55 μm.

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References

1. B. S. Kawasaki, K. O. Hill, D. C. Johnson, and Y. Fujii, Opt. Lett. **3**, 66 (1978).
2. J. Stone, J. Appl. Phys. **62**, 4371 (1987).
3. D. P. Hand and P. St. J. Russell, in *Proceedings of International Conference on Integrated Optics and Optical Fiber Communication* (IOOC, Kobe, Japan, 1989), paper 21C3–4.
4. M. A. Saifi, Y. Silberberg, H. Fouckhardt, A. M. Weiner, and M. J. Andrejco, in *Proceedings of Conference on Lasers and Electro-Optics* (Optical Society of America, Washington, D.C., 1989), paper THH3.
5. Assuming that $P = 100$ mW in the core and an induced loss of $\alpha = 0.23\ \mathrm{m}^{-1}$ (1000 dB/km), the temperature rise in the core would be $(\alpha P/2\pi k)\ln[r_f/w(488)] \sim 0.0016$ K, where $k = 9.2$ W/mK, $r_f = 40\ \mu$m is the fiber outer radius, and $w(488) = 0.76\ \mu$m is the spot radius at 488 nm. Given that $\mathrm{d}n/\mathrm{d}T = 1.3 \times 10^{-5}\ \mathrm{K}^{-1}$, this predicts a thermal Δn of 2×10^{-8}.
6. See L. J. Poyntz-Wright and P. St. J. Russell, Electron. Lett. **25**, 478 (1989), and references therein.
7. M.-V. Bergot, M. C. Farries, M. E. Fermann, L. Li, L. J. Poyntz-Wright, P. St. J. Russell, and A. Smithson, Opt. Lett. **13**, 592 (1988).
8. E. J. Friebele and D. L. Griscom, in *Proceedings of the Materials Research Society Symposium* (Materials Research Society, Pittsburgh, Pa., 1986), p. 319.
9. P. D. Townsend and J. C. Kelly, *Colour Centres and Imperfections in Insulators and Semiconductors* (Chatto and Windus, London, 1973), pp. 35 *ff.*