Light-induced erasure of self-organized $\chi^{(2)}$ gratings in optical fibers

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Self-organized $\chi^{(2)}$ gratings in optical fiber, prepared by irradiating the fiber with 1.06- μ m light from a Q-switched mode-locked Nd: YAG laser simultaneously with its second harmonic, can be erased optically. This is accomplished by irradiating the prepared fiber with the 532-nm mode-locked Q-switched light alone, without the 1.06- μ m light, or with intense green or blue light from an argon laser. The erasure is reversible, and the fiber can be reprepared as before for second-harmonic conversion. The nonexponential time dependence of the erasure process can be explained by a model involving charge separation during seeding and recombination during erasure.

Early observations of second-harmonic generation in optical fibers^{1,2} long seemed at odds with what was thought to be a forbidden or highly inefficient process because of the centrosymmetric, amorphous nature of glass.3 More-recent work4-7 has shown that efficient harmonic conversion in an optical fiber can be achieved at certain wavelengths, particularly if the fiber is irradiated or seeded simultaneously with the fundamental and harmonic light for a few minutes, as reported by Stolen and Tom.⁶ The $\chi^{(3)}$ optical rectification of these two waves induces a periodic $\chi^{(2)}$ in the fiber, with a period equal to the beat length between the fundamental and its harmonic.⁶ The conversion of the fundamental to the second harmonic is efficient because of the long phased-matched interaction length [through the periodic $\chi^{(2)}$] possible in fibers. In the first enhanced doubling experiments of Osterberg and Margulis^{4,5} only fundamental light was launched into the fiber; a weak second harmonic due to the small quadrupole interaction may possibly have sufficed to initiate the process of self-organization.8 Stolen and Tom have shown that seeding the fiber with harmonic light from a doubling crystal along with the fundamental light is a much more effective way to induce selforganization, reducing the preparation time from many hours to a few minutes.⁶ However, the exact nature of the transformation leading to a nonzero $\chi^{(2)}$ inside the fiber is still unclear and subject to some controversy. Some authors have attributed it to the formation of color centers,9 while others have assumed defects or charged traps^{6,10} and stressed the importance of their periodic orientation. Orientation is postulated to be due to the periodic dc electric field arising from the third-order optical rectification of the fundamental and harmonic light.6

We found out that a fiber that was prepared by seeding for second-harmonic generation at 1.06 μ m lost its conversion efficiency after irradiation by the mode-locked Q-switched 532-nm light alone, without the fundamental. Erasure also occurred when the prepared fiber was irradiated with intense green or blue light from a cw argon laser. This erasure effect was also totally reversible, and the fiber recovered the same conversion efficiency if it was reseeded after era-

sure with the fundamental and harmonic light. The study of the dynamics of the erasure brings new insights into the possible mechanisms leading to a non-zero $\chi^{(2)}$.

The fiber used in our experiments was a Corning experimental single-mode fiber with a cutoff at approximately 600 nm. This fiber has a germaniumdoped 3.5- μ m-diameter core. The Δ value is 0.31%, and the molar concentration of germanium is approximately 3%. There is no phosphorus in this fiber. We cut pieces about 35 cm long and seeded them with 1.06-µm light from a mode-locked Q-switched Nd:YAG laser, along with its second harmonic at 532 nm, generated by a temperature-tuned BaNaNbO₃ crystal. The peak infrared power launched into the fiber was as high as 20 kW, although generally it was kept lower to avoid damage to the fiber input end. Seeding the fiber raised the harmonic conversion efficiency rapidly to saturation within a few minutes, as observed first by Stolen and Tom.⁶ The fiber was not single mode at 532 nm, and the green light generated was predominantly in the HE₂₁ mode.

The setup used for the bleaching of the $\chi^{(2)}$ grating is shown in Fig. 1. The Nd:YAG fundamental light was split into two paths by a 50% beam splitter. The light in path 1 was used to probe the conversion efficiency, while path 2 was blocked. The light in path 1 was launched into the prepared fiber, and the generated green light at the output was separated from the fundamental by a polarizing beam splitter. As the seeding green light was polarized at 90° with respect to the fundamental, this orientation was also kept by the prepared fiber. A portion of the green light was reflected onto a glass plate and sent to a photomultiplier tube (PMT). Color filters in front of the PMT further rejected any remaining infrared light, and neutraldensity filters ensured operation of the PMT in a linear region. The light in path 2 was frequency doubled by the same BaNaNbO₃ crystal used for seeding. The generated green light was separated from the fundamental with the same polarizing beam splitter and launched into the prepared fiber through the other end to bleach the $\chi^{(2)}$ grating, while path 1 was blocked. The two paths were alternately blocked or

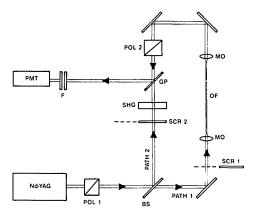


Fig. 1. Setup for bleaching the $\chi^{(2)}$ grating. POL 1 and POL 2, polarizers; BS, beam splitter; SHG, second-harmonic generator; GP, glass plate; MO, microscope objective; OF, optical fiber; SCR 1 and SCR 2, blocking screens; F, filters; PMT, photomultiplier tube.

opened to allow bleaching for a given time and probing of the conversion efficiency, measured as the green average power P_g generated in the fiber. The average power of the fundamental light at 1.06 μ m was kept constant throughout the experiment. At no time did we observe bleaching of the $\chi^{(2)}$ grating by irradiation with the 1.06- μ m light. Measurements of the conversion efficiency as a function of bleaching time t_b were made for different average bleaching powers P_b . The bleaching green power launched into the fiber was changed by slight misalignments of the input microscope objective. Some results are shown in Fig. 2(a). The conversion efficiency was normalized to its value measured immediately after seeding, i.e., at $t_b = 0$.

The decay of the conversion efficiency as a function of bleaching time clearly does not follow a simple exponential law. In fact, it was always observed that the decay rate decreased with increasing time. Assuming that the second harmonic is caused by a $\chi^{(2)}$ grating of constant amplitude A, this behavior could be described by the following decay law:

$$\frac{\mathrm{d}A}{\mathrm{d}t_b} = -\beta(P_b)A^2,\tag{1}$$

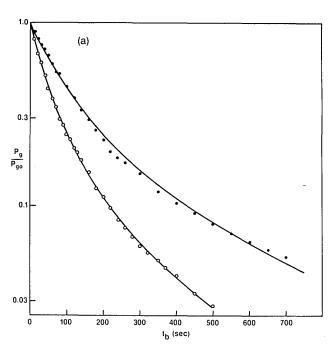
where $\beta(P_b)$ is the decay rate that depends on the bleaching power P_b . If we assume further that the generated green power P_g is proportional to A^2 , as predicted by Stolen and Tom⁶ for P_g much smaller than the seeding green power, then Eq. (1) leads to the following relation for the generated green power as a function of bleaching time:

$$(P_{g0}/P_g)^{1/2} = \beta(P_b)A_0t_b + 1, \tag{2}$$

where P_{g0} and A_0 are the generated green power and the $\chi^{(2)}$ grating amplitude at $t_b=0$, respectively. Plotting $(P_{g0}/P_g)^{1/2}$ as a function of t_b , we found an excellent agreement with Eq. (2), as can be seen from Fig. 2(b). The parameters of the linear-regression fit were also used to draw the theoretical curve in Fig. 2(a). In our experiments the second harmonic was generated mostly in the first few centimeters (<10 cm) of the fiber. Our assumption of a constant grating amplitude and/or period therefore is not strictly valid. Nu-

merical simulations show, however, that the function $(P_{g0}/P_g)^{1/2}$ remains linear even for a nonconstant grating amplitude.

We also measured the decay rate β for four values of P_b and plotted the results on a log-log graph (Fig. 3). A linear-regression fit gave a slope of 4.4 ∓ 1.2 , indicative of the strong power dependence of the bleaching rate. Other sets of data consistently gave an exponent of the order of 3-4 for the bleaching power dependence of β . Some sources of error, however, limit the accuracy of the measurement, namely, the fluctuations in the bleaching power, the fundamental light power, and the possible dependence of β on the model excitation of the bleaching light, which was not accurately con-



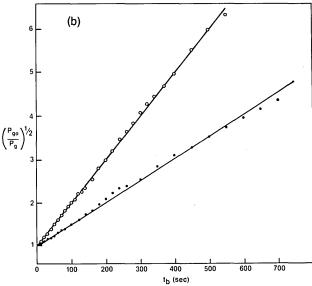


Fig. 2. (a) Normalized average green power generated in the fiber (P_g/P_{g0}) as a function of bleaching time t_b for bleaching powers $P_b = 1.74 \,\mathrm{mW}$ (closed circles) and 2.34 mW (open circles). (b) Same data as in (a) plotted as $(P_{g0}/P_g)^{1/2}$ versus t_b . The straight lines are linear fits whose parameters were also used to draw the curves in (a).

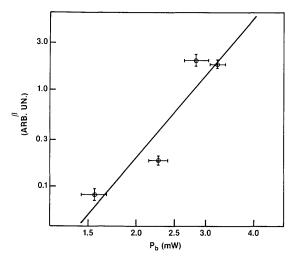


Fig. 3. Bleaching time constant β as a function of bleaching power P_b . The straight line is a linear fit.

trolled. Since we varied the bleaching power by slightly misaligning the input microscope objective, this could have changed the modal content of the bleaching light in the fiber and the value of β . Still, a multiphoton process seems to be responsible for the erasure effect.

Initially we had observed the erasure of the $\chi^{(2)}$ grating by using cw argon laser irradiation of the prepared fibers. In this case, however, the average power was of the order of 600 mW, compared with $\approx 3 \text{ mW}$ of mode-locked Q-switched light at 532 nm for comparable bleaching rates. This experiment was complicated by the fact that the Nd:YAG and argon lasers were not located in the same laboratory, and detailed measurements could not be easily performed. Those initial results, however, indicated that the decay followed the same nonexponential law and was about two times faster with irradiation at 488 nm than at 514 nm. The decay rate also depended strongly on the irradiating bleaching power. The difference in the power levels between cw and mode-locked Q-switched irradiation is also indicative of the multiphoton nature of the process.

One model that may serve to explain the dynamics of the bleaching as given by Eq. (1) is based on charge separation. In this model, charge carriers are excited in the conduction band during seeding, probably ionized from filled traps. These charges (either electrons or holes) move in the dc electric field induced by the mixing of the fundamental and harmonic light and are eventually captured by empty traps in regions of low dc field, where the intensity, and thus the ionization rate, is also lower. The result is an accumulation of charge on each side of the core that itself induces a permanent dc field responsible for the nonzero $\chi^{(2)}$. This model does not exclude contributions to $\chi^{(2)}$ due to orientation of defects in the core by the permanent dc field. During bleaching, the green light ionizes the traps, and the free electrons and/or holes move again in the electric field but now to restore a uniform neutral charge distribution. One can then think of the fiber as a charged capacitor. The charges are made mobile by absorption of light; thus the light changes the effective resistance of the circuit. The current induced will then be proportional to the number of ionized charges, which in turn is proportional to the number of remaining charges. But the current is also proportional to the voltage in the capacitor, which is also in turn proportional to the number of remaining charges. One thus gets the dependence

$$dQ/dt = -kQ^2. (3)$$

This simple model has yet to be confirmed, and the exact nature of the traps has yet to be identified. Many kinds of defects and traps can be found in optical fibers, depending on the dopants used and the fabrication conditions. ¹¹⁻¹⁵

In conclusion, we have shown that the self-organized $\chi^{(2)}$ grating created by seeding an optical fiber with the fundamental and harmonic light of a Nd:YAG laser at 1.06 µm can be erased optically. This is accomplished by irradiating the fiber with the 532-nm light from the mode-locked Q-switched frequency-doubled Nd:YAG laser or with green or blue light from an argon laser. The decay of the conversion efficiency as a function of bleaching time is not exponential but rather follows a law in which the time derivative of the $\chi^{(2)}$ grating amplitude dA/dt is proportional to A². This erasure of the $\chi^{(2)}$ grating is reversible, and seeding the fiber again makes it recover its previous harmonic conversion efficiency. Our results can be explained by a model involving charge separation during seeding and recombination during bleaching. They show that a knowledge of the dynamics of self-organization in the optical fiber is essential to understanding better its exact nature.

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