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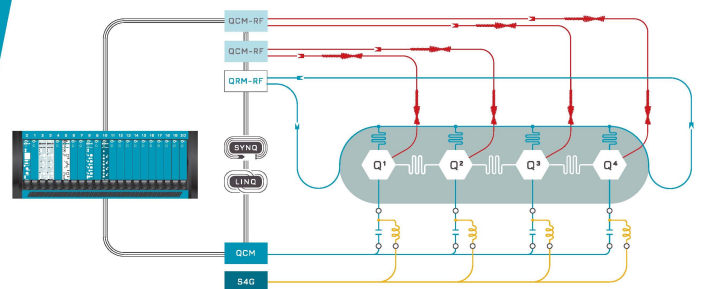
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Light-controlled erasure of induced $\chi^{(2)}$ in thermally poled glasses

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We demonstrate that the induced second order nonlinearity $\chi^{(2)}$ generated in thermal poled doped borosilicate glasses can be erased by a prolonged exposure to near IR laser radiation. The evolution of the second harmonic signal with respect to laser time exposure, intensity and wavelength is reported. The IR laser radiation induced relaxation of the $\chi^{(2)}$ is mainly related to two-photon absorption of the glass. The nonexponential time dependence of the second harmonic signal agrees with a simple model involving diffusion of photoexcited carriers. The reversible erasure process is also observed when the sample is exposed to near ultraviolet laser radiation. © 1999 American Institute of Physics. [S0003-6951(99)03018-1]

Glasses are amorphous materials with inversion symmetry on a macroscopic scale. The inversion symmetry precludes the occurrence of second order optical nonlinear processes. However, Myers, Mukherjee, and Brueck¹ have shown that second harmonic generation (SHG) can be produced in thermally poled fused silica. The thermal poling treatment consists of applying an electric field ($E \sim 3\text{--}5$ kV/cm) below the glass transition temperature ($T \sim 250\text{--}350^\circ\text{C}$) and cooling the glass before removing the dc field. The amplitude of the induced second order nonlinearity $\chi_{\text{ind}}^{(2)}$ observed only near the anodic surface is about 1 pm/V. Other poling techniques have been used and result in similar levels of nonlinearity.^{2,3} However, it has recently been shown that glass fiber poled under UV excitation produces a $\chi_{\text{ind}}^{(2)}$ of about 3.4 pm/V.⁴ In the past seven years, the thermal poling technique has been used to induce a permanent $\chi_{\text{ind}}^{(2)}$ in many glasses.^{5–7} Although the exact nature of the mechanism leading to a nonzero $\chi_{\text{ind}}^{(2)}$ is still unclear, it is generally agreed that two mechanisms, i.e., frozen-in electric field E_0 ($\chi_{\text{ind}}^{(2)} = \chi^{(3)}E_0$ where $\chi^{(3)}$ is the third order nonlinearity of the glass) and oriented dipoles, can explain the nonlinearity measured.^{8,9} The occurrence of $\chi_{\text{ind}}^{(2)}$ in glasses has attracted much interest because it offers the prospect of monolithically integrated electro-optic modulators, switches and frequency converters. For the latter application, a quasiphase matching (QPM) condition must be created. The advantage of QPM in glass waveguides with respect to polar crystal waveguides is mainly related to the lower dispersion of glass waveguides and higher damage threshold. Yet a problem generally encountered in thermal poling is the spreading out of the poled regions due to air breakdown when attempts are made to induce the QPM condition. This problem may be overcome by a selective electron beam erasure or thermal poling in vacuum using patterned electrodes.¹⁰ A collimated light beam may also be used. Indeed, an erasure of $\chi_{\text{ind}}^{(2)}$ in silica submitted to UV laser light has been reported.¹¹ Very re-

cently a decrease in $\chi_{\text{ind}}^{(2)}$ was observed when soft glasses were exposed to intense infrared (IR) laser radiation.¹² Although these experiments show great promise for controlling second order nonlinearity, no systematic study of the influence of the exposure time, intensity and use of wavelength on the $\chi_{\text{ind}}^{(2)}$ erasing process has been performed to date. Moreover, no explanatory mechanisms have been proposed.

This letter is devoted to the study of the evolution of SHG intensity with respect to time exposure, laser intensity and wavelength in Nb₂O₅-doped borophosphate glasses. A nonexponential decay of the second harmonic (SH) intensity with respect to time is observed. A simple model involving the diffusion of photoexcited carriers explains the nonexponential time dependence of the SH intensity.

The samples were 0.8 (0.9 Ca(PO₃)₂-0.1 CaB₄O₇): 0.2 Nb₂O₅ and 0.7 (0.9 Ca(PO₃)₂-0.1 CaB₄O₇): 0.3 Nb₂O₅ described previously.⁷ The glass samples were cut and polished to the dimension of 10×10 mm with a 1 mm thickness. The glass plates were inserted between two planar brass electrodes and heated to about 300 °C. A dc bias of 3 kV was applied across the sample for 1 h. Then the sample was cooled. The dc bias was removed when the sample was at room temperature.

Q-switched (10 Hz repetition rate, 10 ns, 30 mJ) Nd:yttrium-aluminum-garnet (YAG) laser pulses at 1064 nm were used to probe the second order nonlinearity. The pump laser beam polarized in the plane of incidence was focused on the anodic surface using a 10 cm focal length lens. The angle of incidence was chosen to be equal to 45°. The transmitted SH signal was separated from the fundamental radiation using a 1064 nm high reflector, a colored glass filter and a monochromator. The SH signal was detected using a photomultiplier. The linearity of the detection was verified using calibrated optical attenuators. The SH signal was found to increase linearly with the square of the pump pulse energy. The SH signal was stable for more than 1 h.

We performed a similar experiment using a 120 fs continuous wave mode locked Ti:sapphire laser working be-

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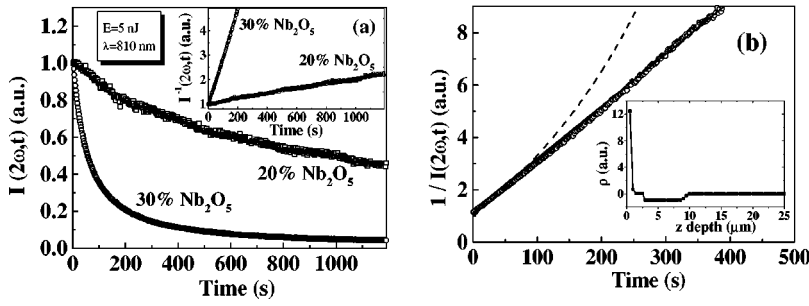


FIG. 1. (a) Temporal evolution of the SH intensity and (inset) inverse of the SH intensity of borophosphate glass doped with 20% (\square) and 30% (\circ) of Nb_2O_5 . (b) Experimental (\circ) and theoretical (solid line: diffusion model and dashed line: diffusion + drift in the built-in field model) evolution of the SH intensity in a borophosphate glass doped with 30% Nb_2O_5 .

tween 800 and 900 nm and yielding pump pulses having about the same peak power. The setup has been described previously.⁸ The SH pulses (<1 fJ) were detected using a photomultiplier coupled to a computer-controlled photon counting system. As shown in Fig. 1(a) the temporal evolution of the p -polarized SH intensity $I(2\omega, t)$ generated by the 5 nJ p -polarized pump pulse centered at 800 nm is quite different. The inset of Fig. 1(a) shows that $I^{-1}(2\omega, t)$ is a linear function of time. We noticed that both the initial SH intensity $I(2\omega, t=0)$ and the slope of $I^{-1}(2\omega, t)$ increased linearly with the square of the pump pulse energy. The latter phenomenon is shown in Fig. 2a. We also recorded the temporal evolution of $I(2\omega, t)$ with respect to the central wavelength of the pump pulse at constant pump pulse energy. Figure 2(b) shows that the decay rate of $I^{-1}(2\omega, t)$ decreased as the Ti:sapphire laser was tuned to shorter wavelengths. The change in the slope is significant, decreasing by two orders of magnitude when the wavelength is shifted from 800 to 900 nm. The decrease of the slope is related to the decrease of the edge absorption band of the sample at the SH frequency [Fig. 2(c)]. This absorption was similar in fresh, poled and depoled samples.

In view of these experiments, it was difficult to conclude whether the recorded $\chi_{\text{ind}}^{(2)}$ erasure was related to one photon near UV absorption or to two photons near IR absorption. To clarify this issue, we performed an experiment where the sample was submitted to two pulses whose central frequencies were, respectively, 800 and 400 nm. The 4 nJ Ti:sapphire pump pulses were split using a mirror ($R=20\%$). The first 3.2 nJ Ti:sapphire pulses were frequency doubled (FD) using a 2 mm potassium dihydrogen phosphate (KDP) crystal cut for type I phase matching. The 20 pJ FD pulses were separated from the fundamental using a colored glass filter. The second 0.8 nJ pulses induced a weak SH signal in the sample studied and were used to check the $\chi^{(2)}$ erasure. These two collinear and overlapping weak pump and FD pulses were focused on the sample. The SH signal was first recorded for a few seconds while the FD pulses were blocked. Then the weak pump pulses were blocked, the SH detection was stopped and the FD pulses were sent through the sample for about 2 min. The procedure was repeated many times. Since the FD pulse energy was a few orders of magnitude larger than the energy of the SH pulse generated in the previous experiment, the erasure process due to near UV absorption had to be speeded up. In fact, the recorded SH signal decay was roughly ten times smaller. We observed that the slope of the $I^{-1}(2\omega, t)$ increased linearly with the FD pulse energy. Notice that the weak pump pulse energy was so small that hardly any decrease of the SH signal was recorded in the sample during the few seconds exposure.

Therefore, this experiment not only shows that near UV light can induce an erasure of the $\chi_{\text{ind}}^{(2)}$ but also indicates that the erasure of the $\chi_{\text{ind}}^{(2)}$ near 800 nm is mainly related to IR two-photon absorption. We performed all these experiments in a 20% Nb_2O_5 doped glass sample and recorded a similar behavior. However, the decrease of the SH signal was less great as a function of time [Fig. 1(a)]. This behavior was correlated with the shift towards the lower frequency of the edge absorption band of this sample [Fig. 2(c)]. Finally in all samples, the erasure was reversible and the glass could be re-prepared as before for SHG by applying the thermal poling treatment.

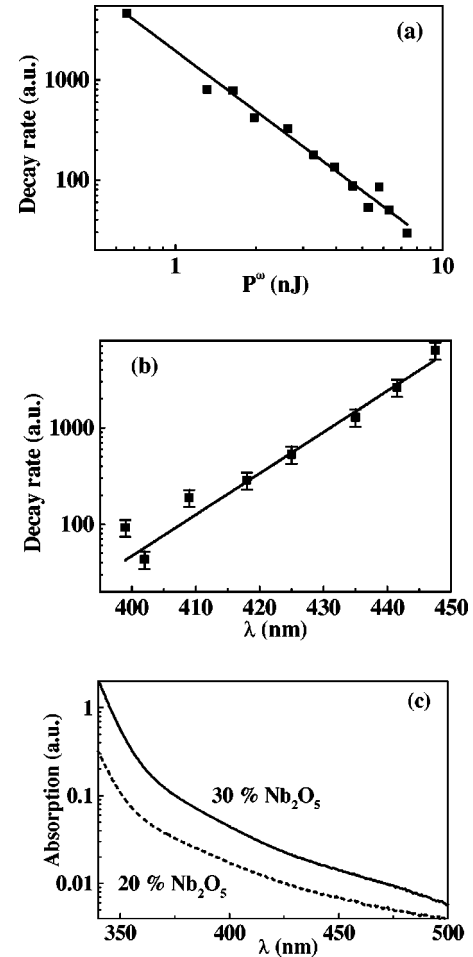


FIG. 2. (a) Evolution of the decay rate of the SH intensity as a function of pump pulse energy at $\lambda=810$ nm. The straight line is a fit of the data assuming the decay rate proportional to the square of the pump pulse energy. (b) Evolution of the decay rate of the SH intensity as a function of the central wavelength of the generated SH pulse for a 5 nJ pump pulse energy. The straight line results from an exponential fit of the data. (c) Absorption of borophosphate glasses doped with 20% (dashed line) and 30% (straight line) Nb_2O_5 .

As previously noted, the decay of the SH signal as a function of time does not follow a simple exponential law. In fact, according to our experimental data, the behavior of the SH signal $I(2\omega, t)$ can be described by the following decay law:

$$\frac{\partial I(2\omega, t)}{\partial t} = -\frac{1}{A}\beta(\omega)I^2(2\omega, t), \quad (1)$$

where $\beta(\omega)$ and A are, respectively, a coefficient which depends on the central frequency of the pump pulses and a constant. Indeed, if one integrates Eq. (1) by taking into account the boundary condition at $t=0$, $I(2\omega, t=0)=AI^2(\omega)$ it is possible to obtain the temporal evolution of the SH signal $I(2\omega, t=0)/I(2\omega, t) = 1 + \beta(\omega)I^2(\omega)t$. This evolution perfectly agrees with our experimental observations.

One model that may help to explain the dynamics of the SH signal is based on the motion of photoexcited carriers. Indeed, it has been shown that at high temperature ($\sim 300^\circ\text{C}$) during the poling process, a large electrostatic field E_0 of the order of 10^7 V/cm is created below the anodic surface by the migration of charges. As the temperature is lowered, the induced charge distribution is frozen and a large E_0 is created in the sample close to the anodic surface.^{1,9} By means of the third order susceptibility $\chi^{(3)}$ this field creates a second order susceptibility: $\chi_{\text{ind}}^{(2)} \sim \chi^{(3)}E_0$. Accordingly, the photoexcited motion of either the trapped charges or neutralizing carriers, respectively, outside or inside this anodic surface layer, lowers E_0 and reduces $\chi_{\text{ind}}^{(2)}$. The photoexcitation of trapped charges or the neutralizing carriers in the conduction band by either one- or two-photon processes is likely. Indeed, a large variation in the erasure process as a function of the wavelength of the pump pulses used is clearly observed. If the wavelength (i.e., $\lambda = 1064$ nm) is situated far from any one- or two-photon absorption bands of the glass, the erasure process is negligible. When the wavelength is tuned to either one- or two-photon edge absorption bands of the sample, the erasure process is clearly activated.

To understand further the $\chi_{\text{ind}}^{(2)}$ erasure process by photoexcitation of neutralizing carriers, we developed a numerical computation where we supposed that the depleted carrier region, created close to the anodic surface during poling, is photoexcited and moves in the volume of the sample according to the equations presented in Ref. 8. A one-dimensional charge distribution was assumed as depicted in the inset of Fig. 1(b). Such a charge distribution has recently been evi-

denced in thermally poled fused silica.¹³ We first neglected the carrier mobility μ . Therefore, as the positive and negative photoexcited charges diffuse, they neutralize each other, reduce the amplitude of the static build in electric field $E_0(z, t)$ and induce a decrease of the SH signal. The computed temporal evolution of the resulting second order signal

$$I(2\omega, t) = \left[\int_0^\infty E_0(z, t) dz \right]^2$$

is reported in Fig. 1(b) (solid line). It is interesting to note that such a simple model agrees very well with the experimental data. Another model was considered in which both the mobility and the diffusion of these photoexcitable trapped charges were taken into account. In this way, they could also drift within the built-in electric field $E_0(z, t)$. The evolution of the resulting signal $I(2\omega, t)$ is presented as a dashed line in Fig. 1(b). Note that, contrary to our previous result, the computed behavior did not fit the experimental data. Therefore, our results seem to indicate that the motion of the photoexcited carriers is mainly driven by diffusion.

In conclusion, we show that the $\chi_{\text{ind}}^{(2)}$ generated in thermal poled doped borophosphate glasses can be erased by a prolonged exposure to near IR laser radiation. The relaxation of $\chi_{\text{ind}}^{(2)}$ induced by IR and near UV laser radiations is related to two- and one-photon absorption, respectively. The time dependence law of the erasure process is explained by a simple model involving a diffusion of photoexcited carriers.

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