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Linear and second order nonlinear gratings induced by two photon absorption in thermally poled glasses.

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Non phase matched second harmonic generation (SHG) can be produced in thermally poled fused silica [1]. The second order susceptibility $\chi_{ind}^{(2)}$ induced over $\sim 10 \mu\text{m}$ beneath the anodic surface is due to coupling of the third order glass susceptibility $\chi^{(3)}$ with a built in electric field E_{int} ($\chi_{ind}^{(2)} \sim \chi^{(3)} E_{int}$). To achieve quasi-phase matching (QPM) conditions, different techniques have been proposed and a 20% SHG efficiency has been demonstrated in QPM thermally poled fibers [2]. Here using amplified femtosecond pulses in thermally poled glasses, we propose a new method which results within a couple of minutes in the formation of both thick linear and non linear QPM thin gratings. Our results also show that thermally poled glasses behave as erasable optical memories.

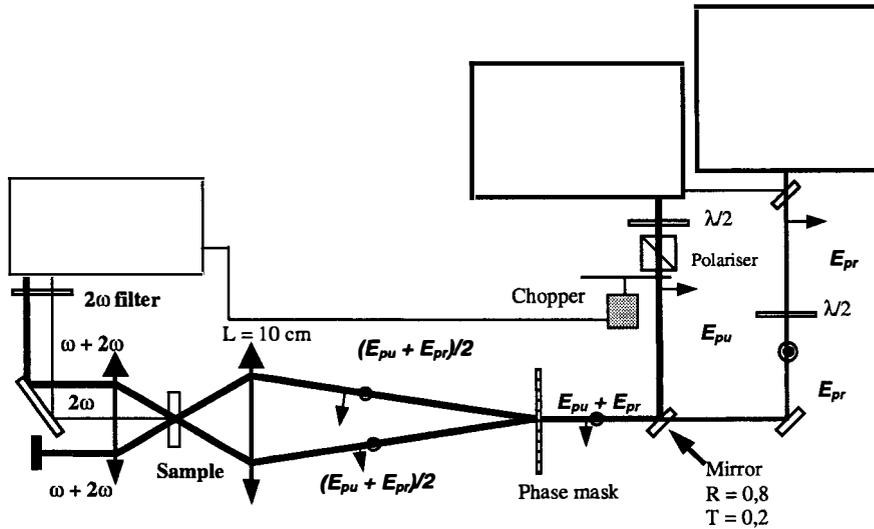


Figure 1: *Experimental set-up*

The experimental set-up, presented in Figure 1, uses femtosecond non amplified (76 MHz C.W mode-locked, 120 fs, 10 nJ) and amplified (1 kHz, 0.5 mJ) Ti: Sapphire laser pulses centered at 810 nm as probe and pump beams, respectively. The collinear pump and probe pulses, linearly and orthogonally polarized respectively, were sent through a phase grating (333 grooves/cm). After the grating the two equally balanced and focussed ($w = 160 \mu\text{m}$) pump beams were overlapped and produced a periodic intensity pattern ($I(r) = I_0(1 + \cos(2\pi.r/\Lambda))$ where $\Lambda = 3.3 \mu\text{m}$) in the thermally poled 30% Nb_2O_5 doped borophosphate glass sample. The samples were poled at 300°C for about 1 hour applying a dc bias of 3 kV

across the 1 mm thick glass. They were tested using weak probe beams ($E_{pr} \sim 0.5$ nJ). With a single probe beam going through the sample, steady SHG was recorded along the beam direction, whereas with the two synchronized and overlapping probe beams, SHG was detected in both the probe beams and in the non-collinear SHG direction. Then, the pump beam modulated by a mechanical chopper was sent through the sample. The temporal evolution of the probe SHG recorded when the pump beam was blocked, is presented in Figure 2. These figures demonstrate the pump beams erase the $\chi_{ind}^{(2)}$. (Fig. 2 a-b). The temporal decay of the SHG signal $S(r, 2\omega, t)$ well agree with a one dimension spatial extension of the decay law, we recently proposed [3] (Fig. 2a-b full-line). Indeed, according to the two photon-erasure process one can consider that the time dependent SHG signal evolve as:

$$\frac{S(r, 2\omega, t=0)}{S(r, 2\omega, t)} = \left(\frac{\chi_{ind}^{(2)}(r, t=0)}{\chi_{ind}^{(2)}(r, t)} \right)^2 = 1 + \beta(\omega) I^2(r, \omega) t \quad (1)$$

where $I(r, \omega)$ describe the spatial profil of the erasure intensity pattern, $\beta(\omega)$ is a coefficient which depends on the two photon absorption of the used glass and $\chi_{ind}^{(2)}(r, t)$ describe the spatial and temporal evolution of the induced second order nonlinearity during the erasure process. Note that at the beginning of the erasure process, we verified $S(r, 2\omega, t=0) \propto (\chi_{ind}^{(2)}(r, t=0))^2$ was almost constant over the whole thermally poled region. We also tried to evidence the formation of a thin non-linear grating $\chi_{ind}^{(2)}(r)$. After the erasure, if one shines this sample with a single probe laser beam one would expect to record SHG along different directions. Indeed, one can easily show that the far field second harmonic intensity pattern generated inside the thin poled region by the spatially modulated $\chi_{ind}^{(2)}(r, t)$ is proportional to the square modulus of the spatial Fourier transform of $\chi_{ind}^{(2)}(r, t)$. In figure 2c we have reported the evolution of the intensity ratio of harmonic generation along the first and second order of diffraction of the induced nonlinear grating. One can note that while the trend is well described the experimental ratio is down shifted compared to the theoretically expected ratio. We believe this clearly indicate the limitation of the one spatial dimension model we proposed.

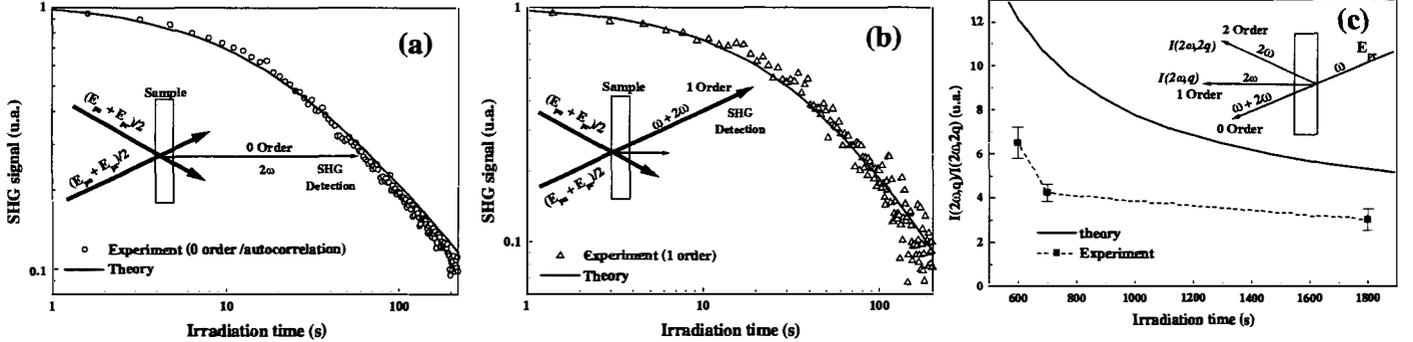


Fig. 2 : a-b: Theoretical (full line) and experimental evolution of the maximum of the intensity autocorrelation (O) and of the first diffraction order (Δ) of the probe beams during second order nonlinear grating formation. c: Theoretical (full line) and experimental (dotted line) evolution of the ratio between the second harmonic intensity recorded in the first and second order Bragg direction. To record the signal the pump is blocked and a single probe beam is sent through the sample.

The diffusion of carriers created by either one or two-photon absorption of the pump beams should reduce E_{int} and may explain the SH signal relaxation [3] or the increase of the $\chi_{ind}^{(2)}$ in UV-excited poled fibers. This might be associated with the formation of photo-refractive linear index grating in the whole sample volume. To evidence this, an experiment was performed using an He-Ne laser beam entering the glass sample at a Bragg diffraction angle. The temporal evolution of the diffracted He-Ne beam is

presented in Figure 3. When the pump beam was on, its intensity rapidly increased and saturated. Its intensity decreases slowly when the pump beam was off. Under this experimental condition we only record a light diffraction along the Bragg direction. We have not been able to observed any increase of the sample absorption after the erasure process. These results clearly indicate we are recording the formation and relaxation of a thick linear index grating. The power dependence of the grating reflectivity (i.e. $\eta \sim I_{pu}^4$, Fig. 2b) [4] and the formation ($\tau \sim I_{pu}^{-2}$ Fig. 2c) [5] time agree with a two-photon photo-refractive phenomenon. We evaluated the maximum index change to be: $\Delta n \sim 5 \cdot 10^{-5}$. Similar thick index grating was also recorded in a non-poled sample.

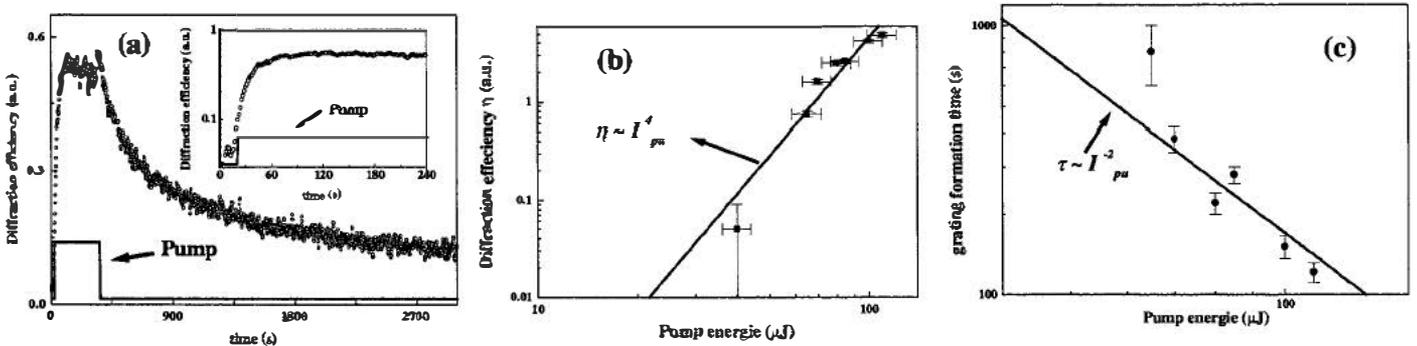


Figure 3. a: Formation and relaxation of linear index grating. **b:** Evolution of steady state grating reflectivity η as a function of pump energy. **c:** Evolution of grating formation time τ as a function of pump energy

Finally, our results also show that thermally poled glasses can be used as erasable optical memories: one can consider that the spatially limited optical eraser of the induced second order non linearity allows to store a bit of information that can be retrieved looking for the absence of SHG at the considered point. In fact, we have shown that poled glasses have less binary level of erasure and have photographic film like behavior.

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