HAL
open science

Quasi phase matched chi(2) gratings printed with various memory mechanisms by dual frequency interferences in polymer films

Raynald Seveno, Aleksandra Apostoluk, Vincent Boucher, Celine Fiorini Debuisschert, P. Raimond, Jean-Michel Nunzi, B. Sahraoui

To cite this version:

Raynald Seveno, Aleksandra Apostoluk, Vincent Boucher, Celine Fiorini Debuisschert, P. Raimond, et al.. Quasi phase matched chi(2) gratings printed with various memory mechanisms by dual frequency interferences in polymer films. Optoelectronics 2003, Jan 2003, San Jose, United States. hal-01512508

HAL Id: hal-01512508

https://hal.science/hal-01512508v1

Submitted on 16 Jan 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Quasi-phase-matched gratings printed by all-optical poling in polymer films

Aleksandra Apostoluk, David Chapron, Grégory Gadret, Bouchta Sahraoui, and Jean-Michel Nunzi,

Laboratoire des Propriétés Optiques des Matériaux et Applications, Unité Mixte de Recherche– Centre National de la Recherche Scientifique 6136, Université d'Angers, 2 Boulevard Lavoisier, 49045 Angers, France

Corresponding author : jean-michel-nunzi@univ-angers.fr

Céline Fiorini-Debuisschert, and Paul Raimond

Commissariat à l'Energie Atomique, Direction de la Recherche Technologique, Laboratoire d'Intégration des Systèmes et des Technologies, Département d'Elaboration et de Contrôle des Structures – Service d'Elaboration et de Mise en Forme des Matériaux, Laboratoire Composants Organiques, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette, France

Abstract

The all-optical poling technique permits polar orientation of molecules. For efficient poling of thin films the relative phases, amplitudes, and polarizations of the two interfering beams must be controlled. We present an original stable one-arm interferometer that is specific to the recording of two-color interference. It relies on the index dispersion of optical glasses. This interference technique permits true real-time nonperturbative monitoring of the polar orientation process and easy all-optical poling of thin-film materials without the need for phase control. This new configuration opens the door to the realization of customized phase-matched wave-guided frequency-conversion devices for the near infrared.

Self-induced generation of second-harmonic light was observed first in an optical fiber illuminated with intense light at 1.06 μm [1] and later with two beams at fundamental (1.06- μm) and second-harmonic [2] (0.532- μm) frequencies, revealing the way to induce second-order nonlinear susceptibility $\chi^{(2)}$ by purely optical methods in a centrosymmetric material. Later, the optical induction of noncentrosymmetry in azo-dye materials has been the object of extensive studies [3]-[6] owing to the larger nonlinearities and potential applications of these materials in integrated optoelectronic devices. Side-chain polymer matrices containing organic moieties with large second-order polarizabilities β indeed combine the possibility of combining large nonlinearities and ease of processing of polymers. Controlled spatial structuring of the nonlinear optical susceptibility on a wavelength scale permits phase matching of light waves at arbitrary frequencies, which may lead to the development of new technologies for the fabrication of integrated tunable sources [7].

The interference between a fundamental wave E_ω and its second harmonic (SH) $E_{2\omega}$ leads to the presence of a polar field $E(t)=E_\omega(t)+E_{2\omega}(t)$ inside the medium. The temporal average of the field cube $\langle E^3(t)\rangle_t$ is nonzero, which results in the selective polar excitation of the molecules at the fundamental and the SH frequencies and in a quasi-permanent polar orientation of the dye molecules, thus breaking the centrosymmetry of the material and resulting in the recording of a $\chi^{(2)}$ susceptibility with a spatial period that satisfies the phase-matching conditions for second-harmonic generation (SHG) [8]. The influence on the efficiency of all-optical poling of such seeding parameters as the relative phase between writing beams, their relative intensities, and the beam polarization states as well as the temperature dependence has been studied by several research groups [9]-[12]. The relative phase is an important factor, particularly when the writing beams are linearly polarized. Efficient SHG requires phase matching, so the harmonic fields generated in different regions along the material interfere constructively at the output. In this Letter we demonstrate a new all-optical poling scheme in which phase is automatically adjusted. The technique also permits real-time monitoring of the induced nonlinearity in thin films.

Samples were thin films of the copolymer DR1-MMA 35/65 described in Ref [3]. Its glass-transition temperature is 130°C. We prepared thin films by spin coating a 50-g/L solution of the copolymer in 1,1,2-trichloroethane onto glass substrates. The film thickness was 0.47 μm, and the optical density at 532 nm was 1.17. The refractive-index difference between the fundamental wave at 1.06 μm and the SH wave at 532 nm, $\Delta n=n_{2\omega}-n_\omega$, was 0.3, and the coherence length, $l_c=\lambda_{2\omega}/\Delta n$, was 1.7 μm [5]. The source was a nanosecond Q-switched Nd3+:YAG laser operating at the fundamental wavelength (1.06 μm). The laser delivered pulses of 7-ns width at a repetition rate of 10 Hz. A type II KDP crystal was used as a frequency doubler. The pulse energy was 20.2 mJ at 1.06 μm and 12.9 μJ at 532 nm. The writing beam's power ratio was optimized to produce the maximum seeded SHG efficiency. The two writing beams (Fig. 1) were copropagating and were weakly focused by a lens (L) onto the sample into a spot of 3-mm diameter. A polarizer (P) was used to ensure parallel polarization of the two beams. The photoinduced second-order susceptibility $\chi^{(2)}$ was probed by SH generation inside the sample and was detected by a photomultiplier tube (PM). The relative phase between the writing beams at frequencies ω and 2ω could be tuned by translation of a wedge-shaped BK7 glass plate with a known index of dispersion. During the readout phase a red-glass filter could be inserted just after the laser output to block the 532-nm light. A filter for the ω beam was placed in front of the photomultiplier tube to stop the infrared beam. A shutter set in front of the photomultiplier and kept closed during the seeding period protected the photomultiplier from damage. The signal from the photomultiplier-tube was measured with a digital oscilloscope (OSC). The sample was placed on an isosceles glass prism with an acute angle of 21.6°. The prism's refractive indices for both fundamental and SH waves were measured by the beam refraction method and were 1.560 and 1.593, respectively.

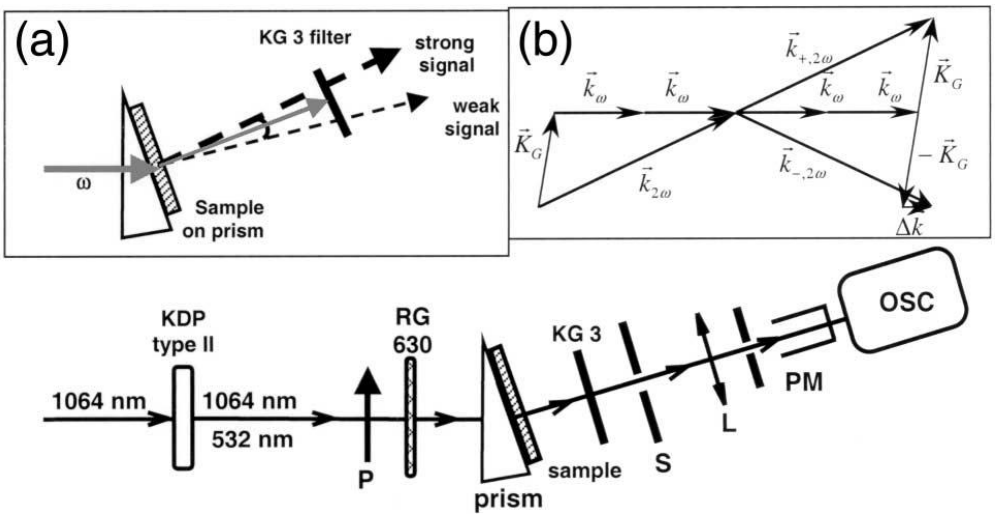

Fig. 1 Experimental setup: RG 630, red Schott filter for the 2ω beam; KG 3, Schott filter for the ω beam. A BK7 wedge-shaped glass plate could be placed just after the KDP crystal to adjust the relative phase between the two writing beams. S, shutter. (a) Beam configuration at the output of the prism;
(b) schematic of wave-vector conservation of the interacting fields.

Seeding was performed for 30 min, the time necessary to achieve saturation of the SH signal. The experiments were performed for samples placed perpendicular to the writing beams (without prism) and for samples placed in contact with the glass prism. In the first case we observed a variation of the generated SH intensity as a function of the relative phase difference, in a similar way as was reported in Ref. [5]. In the second case of a sample placed on a prism [Fig. 1, inset (a)], the SHG signal was independent of the BK7 wedge plate position, as we explain below. The irradiating copropagating ω and 2ω beams were perpendicular to the front side of the prism. We observed two green signals behind the prism: a strong one, which was refracted in the same direction as the 2ω writing beam (during the seeding phase), and a second signal, which was weaker than the first. These two beams were emitted symmetrically on both sides of the fundamental beam, at the output of the prism. This configuration is shown in inset (a) of Fig. 1. The stronger signal observed during the readout period was phase matched transversally and longitudinally. The second one, weaker, was only transversely phase matched, which is the reason for its smaller intensity. The wave vectors of the interacting waves are shown schematically in inset (b) of Fig. 1. Wave-vector conservation gives

$$\mathbf{k}_{+,2\omega} = \mathbf{k}_{\omega} + \mathbf{k}_{\omega} + \mathbf{K}_G = \mathbf{k}_{2\omega}, \qquad (1)$$

$$\mathbf{k}_{-,2\omega} = \mathbf{k}_{\omega} + \mathbf{k}_{\omega} - \mathbf{K}_G + \Delta\mathbf{k} \qquad (2)$$

where $\mathbf{K}_G=\mathbf{k}_{2\omega}-2\mathbf{k}_{\omega}$ is the grating vector, $\mathbf{k}_{\omega}$ is the wave vector at the fundamental frequency, $\mathbf{k}_{+,2\omega}$ is the wave vector at the SH frequency of the strong signal, $\mathbf{k}_{-,2\omega}$ is the wave vector at the SH frequency of the weak signal, and $\Delta\mathbf{k} = 2\pi(n_{2\omega}-n_{\omega})/\lambda_{2\omega}$ is the wave-vector mismatch of the weak signal that occurs in the direction normal to the sample surface.

The presence of a weaker signal permits real-time monitoring of the all-optical poling process. In Fig. 2 we present the dynamics of the all-optical poling process. The SHG signal is monitored in two ways: in seeding–readout sequences, in which the green beam is cut off during the readout process, and in real time. The fast decay observed after the 2ω seeding beam is stopped is due to the relatively large intensity of the probe beam at fundamental frequency (it is equal to the writing fundamental beam's intensity), which destroys orientation [12]. The 2ω decay is the same when $\chi^{(2)}$ is constructed in regular seeding–readout sequences or in real time (Fig. 2, inset), as there is no difference between these two methods when decay is tested. Real-time monitoring prevents partial destruction of the

induced polar orientation when the seeding period is stopped at regular time intervals for probing the encoded $\chi^{(2)}$ while the strong fundamental beam irradiates the sample in the probing step. The intensity of the weak signal measured in real time is proportional to the intensity of the strong signal, and thus we get an insight into the real growth dynamics of the induced $\chi^{(2)}$, not influenced by any partial erasure by the ω beam during the probing step, as is shown in Fig. 2: For the first 15 min of the seeding phase the intensity of the weak signal measured in the real-time regime is clearly higher than the intensity measured in writing–probing sequences. As all-optical poling of bulk glass was performed by several research groups [13] we probed seeding of the prism after 2 h to see an eventual contribution to the SH signal emerge from the prism. No SH signal was observed in such conditions, so the two 2ω beams observed in the experiment with the sample on a prism are attributed to the polymer film.

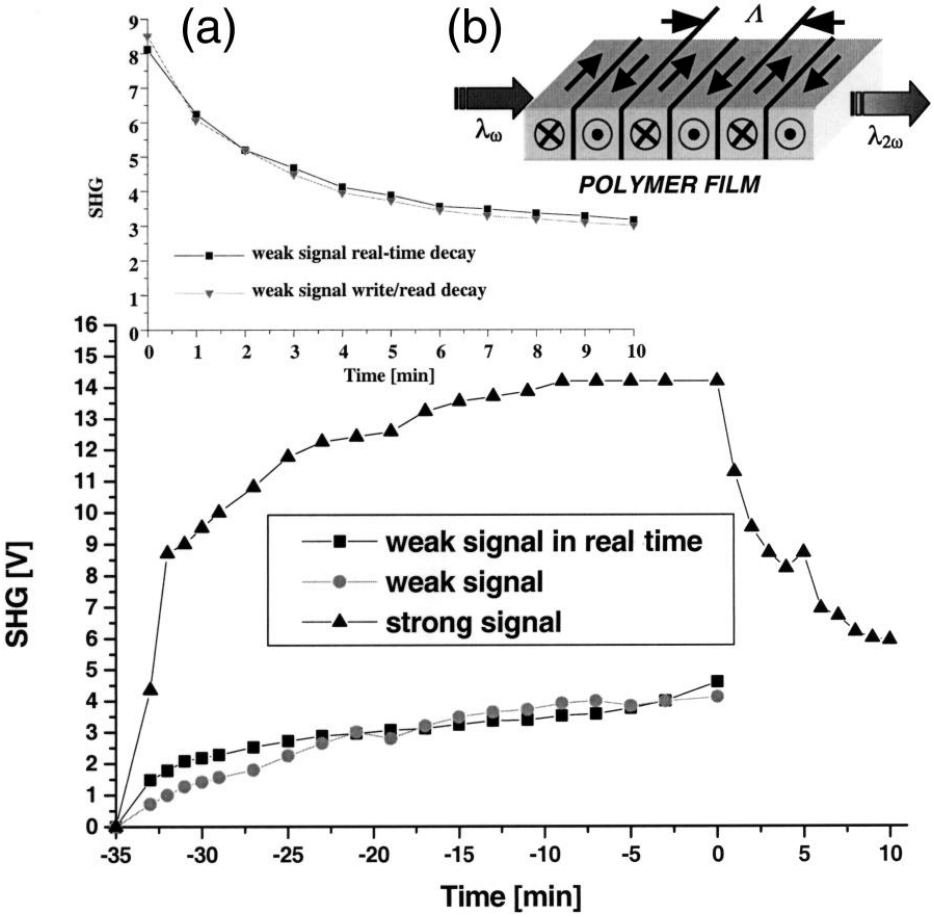

Fig. 2 Real-time growth and decay of the SH-generated signal (in volts). Negative times correspond to seeding preparation; at time zero, seeding has stopped. Positive times correspond to the temporal decrease of the induced $\chi^{(2)}$ susceptibility. The sample was placed on the prism. (a) Decay of the weak signal monitored after seeding under real-time and regular write–read conditions, on different scales. (b) Schematic of the quasi-phase-matched structure induced inside the polymer thin film for use as a frequency-doubling waveguide device. Λ is the period of the inverted domain grating.

Changing the relative phase difference between the two beams can strongly influence the intensity of the SH signal [5]. Indeed, by moving a BK7 plate we can change the position of the fringes on the sample. As the fringe pattern in the air is sparse, moving the fringe pattern may result in a situation in which the sample is put onto a dark fringe, such that no poling is achieved. For the two writing beams interfering inside the prism, the refractive-index dispersion for the two writing waves gives 65 pairs of fringes per millimeter. As the beam diameter is 3 mm, if the sample is placed on a prism we obtain a dense fringe pattern. In consequence, changing the relative phase between the writing beams only

translates the pattern, keeping the number of fringes over the beam diameter almost constant. The harmonic signal observed at the output is thus a constant, with the phase difference between the two seeding beams. When the sample was placed on the prism for seeding, we observed a lower signal than for a free-standing sample with optimized phase. The reason for this is that there are many grating fringes over the beam diameter (65 per millimeter), so the signal is an average of what we would get by varying the phase with a free-standing sample (a factor of 4 is expected theoretically). In the direction parallel to the plane of the sample we obtained a modulated quasi-phase-matched structure with domain inversion [Fig. 2, inset (b)]. It can be used for phase-matched frequency conversion in a planar waveguide configuration [14],[15]. The periodicity of the arrangement depends on the wavelength of the writing beams and on the acute angle of the prism.

The polymer film was removed from the prism after a 30-min seeding period. Diffraction from a 632.8-nm He–Ne laser at normal incidence upon the sample was investigated. No diffraction could be observed, showing that there was no efficient refractive-index grating. There is indeed, in principle, no difference in refractive index between domains where azo molecules are oriented positively or negatively. So we saw no periodic modulation of the birefringence, in contradiction to recent observations of Tsutsumi *et al* [16]. But this result does not prove that birefringence is negligible, as was the case in the real-time monitoring experiment proposed by Churikov *et al* [17]. We monitored harmonic diffraction after 30 min of seeding performed with a sample on a prism. The sample was removed from the prism and was irradiated with the fundamental frequency beam. We observed two weak frequency-doubled signals placed symmetrically about the central fundamental beam [Fig. 1, inset (a)]. We could see that +1 order harmonic diffraction is weaker than −1 order, but we could not measure their intensities under such conditions. The measured angle between the two diffracted harmonic signals was ∼4∘, corresponding to a grating period of ∼17 μm. This gave ∼58 pairs of fringes per millimeter, in good agreement with the calculation made by use of the prism indices and an acute angle.

In conclusion, we have investigated new aspects of the holographic nature of all-optical poling in thin polymer films. We demonstrated that placing the sample upon a glass prism results in a phase-independent all-optical poling process, which is related to the peculiar nature of two-color holography. Thus we found a new technique with which to optimize the phase difference for all-optical poling of thin films. This new method permits true real-time monitoring of the all-optical poling process in all kinds of samples. It could even permit the study of unstable samples. It is also a new processing technique for efficient wave-guided frequency-conversion devices.

References

1. U. Österberg and W. Margulis, Opt. Lett. **11**, 516 (1986).

2. R. H. Stolen and H. W. K. Tom, Opt. Lett. **12**, 585 (1987).

3. F. Charra, F. Kajzar, J. M. Nunzi, P. Raimond, and E. Idiart, Opt. Lett. **18**, 941 (1993).

4. C. Fiorini, F. Charra, J.-M. Nunzi, I. D. W. Samuel, and J. Zyss, Opt. Lett. **20**, 2469 (1995).

5. C. Fiorini, F. Charra, J.-M. Nunzi, and P. Raimond, J. Opt. Soc. Am. B **14**, 1984 (1997).

6. X. Yu, X. Zhong, Q. Li, S. Luo, Y. Chen, Y. Sui, and J. Yin, Opt. Lett. **26**, 220 (2001).

7. C. Fiorini, J.-M. Nunzi, F. Charra, and P. Raimond, Photon. Sci. News **3**(1), 16 (1997).

8. N. B. Baranova and B. Ya. Zeldovich, JETP Lett. **45**, 717 (1987).

9. X. Yu, X. Zhong, Q. Li, S. Luo, Y. Chen, and Y. Sui, Opt. Commun. **180**, 373 (2000).

10. S. Brasselet and J. Zyss, J. Opt. Soc. Am. B **15**, 257 (1998).

11. A.-C. Etilé, C. Fiorini, F. Charra, and J.-M. Nunzi, Phys. Rev. A **56**, 3888 (1997).

12. F. Fiorini and J.-M. Nunzi, Chem. Phys. Lett. **286**, 415 (1998).

13. T. J. Driscoll and N. M. Lawandy, J. Opt. Soc. Am. B **11**, 355 (1994).

14. S. Somekh and A. Yariv, Opt. Commun. **6**, 301 (1972).

15. M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, IEEE J. Quantum Electron. **28**, 2631 (1992).

16. N. Tsutsumi, J. Yamamoto, and W. Sakai, Jpn. J. Appl. Phys. **40**, 2264 (2001).

17. V. Churikov, M. F. Hung, and C. C. Hsu, Opt. Lett. **25**, 960 (2000).