Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Modulation of the Optical Second Harmonic Generation in Ferroelectric Liquid Crystals

Irena Drevenšek [a], Renato Torre [b] & Martin Čopič [a c]

[a] J. Stefan Institute, University of Ljubljana, Jamova 39, 61111, Ljubljana, Slovenia

[b] European Laboratory for Nonlinear Spectroscopy, Largo E. Fermi 2, 50125, Florence, Italy

[c] Department of Physics, University of Ljubljana, Jadranska 19, 61111, Ljubljana, Slovenia

Version of record first published: 24 Sep 2006.

To cite this article: Irena Drevenšek , Renato Torre & Martin Čopič (1994): Modulation of the Optical Second Harmonic Generation in Ferroelectric Liquid Crystals, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 251:1, 101-109

To link to this article: http://dx.doi.org/10.1080/10587259408027196

Mol. Cryst. Liq. Cryst. 1994, Vol. 251, pp. 101–109
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

MODULATION OF THE OPTICAL SECOND HARMONIC GENERATION IN FERROELECTRIC LIQUID CRYSTALS

IRENA DREVENŠEK*, RENATO TORRE#, and MARTIN ČOPIČ*,"

* J. Stefan Institute, University of Ljubljana, Jamova 39, 61111 Ljubljana, Slovenia
# European Laboratory for Nonlinear Spectroscopy, Largo E. Fermi 2, 50125 Florence, Italy
" also Department of Physics, University of Ljubljana, Jadranska 19, 61111 Ljubljana, Slovenia

Abstract We investigated the temporal dependence of the second harmonic generation from a 50 μm thick homeotropically aligned cell of ferroelectric liquid crystal SCE9 (BDH Ltd.) exposed to a square waveform field varying between non zero and zero value. The field switches the molecular configuration between the efficient phase matching geometry and the inefficient helically modulated structure. The power of the second harmonic beam responds to the field on a millisecond time scale. The response time of the signal after switching on the field decreases inversely proportionally to the amplitude of the field, while the response time of the signal after switching off the field is almost field independent and has a value characteristic for the spontaneous relaxation of the helical SmC* phase (Goldstone mode). The angular dependence of the modulated second harmonic intensity is almost the same as in the static external field. This shows that a completely unwound homogeneous molecular structure is formed in a few ms after application of the field.

INTRODUCTION

A combination of large second order nonlinear optical susceptibility $\chi^{(2)}$ and fast electrooptic response of novel ferroelectric liquid crystals (FLC) makes these materials very convenient for modulation of the optical second harmonic generation (SHG) [1, 2]. This feature opens up a new field of applications in nonlinear integrated optics from simple elements like nonlinear optical switches to complex nonlinear wave guiding units [3]. A fast switchable SHG device may be used for example as a nonlinear intracavity dumper in the laser systems. The FLC materials presently available do not yet adequately meet the demands for this kind of elements.

The most effective modulation of the energy conversion from incident to second harmonic beam in the FLC cell is achieved by switching the molecular configuration into and out from the phase matching geometry. For example the phase matching is lost when the external electric field is removed from the cell with the homogeneous unwound SmC* structure and the helically modulated SmC* form appears. The spatial inhomogeneity of the optical properties in the helically modulated SmC* phase drastically decreases the second harmonic signal [4, 5].

The temporal behaviour of the SHG after application or removal of a stepwise electric field in a thick homeotropic FLC cell was first measured by Ozaki and Yoshino [6]. They observed the response on two characteristic time scales, one in the region of a few milliseconds and another in the region of a few seconds. In this paper we present the results of detailed study of this type of SHG modulation, including the temporal dependence of the second harmonic intensity at different frequencies and amplitudes of the square wave form field and also the angular dependence of the modulated second harmonic signal.

EXPERIMENT

The compound used in our investigation was a commercial BDH mixture named SCE9. It has a room temperature SmC* phase with the tilt angle θ of 20.5° at 20°C. The components of the nonlinear susceptibility χ_{ijk} and the color dispersion of the refractive index of SCE9 have been previously determined by J. Y. Liu at al. [7]. This allowed us to concentrate directly on the dynamic properties of the SHG in this material. A homeotropic sample (with the smectic planes parallel to the bounding planes) was prepared using silane coated glass plates separated by two 50 μm thick mylar spacers. One of the glass plates was covered also by two stripes of conducting ITO layer which served as electrodes to provide electric field parallel to the glass surfaces. The gap between the electrodes was 1.4 mm.

The fundamental light source in our experimental setup was a Q-switched Nd-YLF laser operating at a repetition rate of 1 kHz. It provided 7 ns long light pulses with the 1.046 μm wavelength. The average power of the outgoing beam $\langle P_\omega \rangle$ was 150 mW. The beam passed a polarizer and a visible-cut filter and was than focused on the sample by a cylindrical lens with a focal length of 500 mm. The laser spot size on the sample was around 0.6 mm x 0.1 mm with the long axis parallel to the electrodes. According to the material parameters and sample thickness, this arrangement results in a maximal possible power conversion ratio $P_{2\omega}/P_\omega \sim 5 \cdot 10^{-8}$. The second harmonic beam was separated from the fundamental beam by a grating spectrometer and detected by a

photomultiplier. The output from the photomultiplier was sent to a gated integrator synchronized with the laser. The integrated signal was monitored on a digital oscilloscope where it was averaged up to 256 times. As there was no synchronization between the laser QS unit and the square waveform field generator which was modulating the sample and also triggering the scope, the multiple averaging of the signal provided random sampling giving an effective time resolution of about 10 μs.

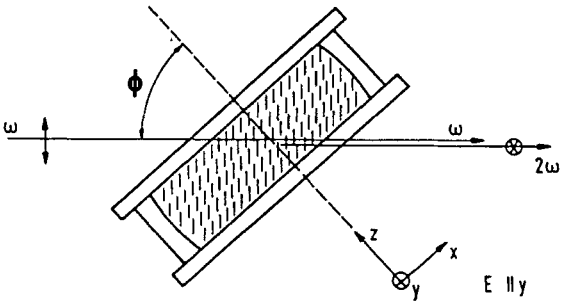

FIGURE 1 Experimental geometry.

All the measurements were performed at room temperature. The local photothermal heating of the sample by the fundamental laser beam was neglected, because it did not cause any significant changes of the optical properties of the sample. This was verified by testing the variation in the polarization of a weak He-Ne laser beam which was passing the same region of the sample than the YLF beam. Almost no difference in the outgoing polarization was detected when the YLF beam was switched on or off. The sample was mounted on the rotation stage which allowed rotation of the sample around the direction of the external field (Fig. 1). An unwinding DC voltage of 220 V was applied to the sample and the position corresponding to the type I (ee-o) phase matching was found. The phase matching was achieved when the incident angle ϕ of the laser beam was 2^{o} with respect to the sample normal. After this the field was changed to the square waveform field.

RESULTS AND DISCUSSION

In the first series of measurements we investigated the dependence of the SHG response on the frequency of the modulating voltage. The amplitude of the applied voltage was 220 V. This corresponds to the field about seven times larger than the critical field E_c necessary to unwind the helical structure. Figure 2 shows the temporal

behaviour of the second harmonic power $P_{2\omega}$ at four different frequencies of the modulating field. The temporal dependence of the field is also marked. At the frequencies below 1 Hz $P_{2\omega}$ almost ideally follows the field. This is contrary to the significant slow response reported in ref. [6]. In our experiments, we only observe a very small amount of the signal which decays on a second time scale after the field is switched off. At a bit higher frequencies the delay of a few ms after switching on the field and the delay of a few tens of ms after switching off the field become evident. At even higher frequencies the modulation is becoming less and less efficient and finally at the frequency around 50 Hz it disappears. At 50 Hz the molecules do not react to the field steps anymore so that the structure is permanently unwound and the SHG is present all the time.

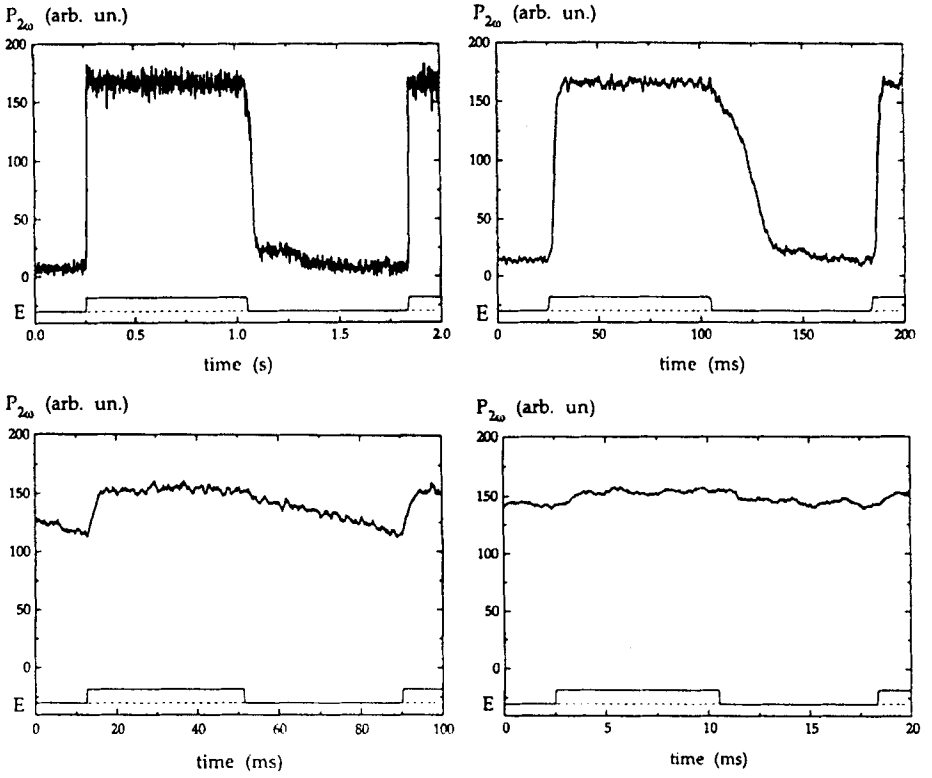

FIGURE 2 Temporal dependence of the SHG at different frequencies of the modulating square waveform field corresponding to the applied voltage with the amplitude of 220 V. Temporal dependence of the field is shown at the bottom of the figures. The dashed line corresponds to zero field.

The limiting modulation frequency of 50 Hz is relatively low for applications and the first idea to increase it is to increase the amplitude of the modulating field. Figure 3 shows the temporal behaviour of $P_{2\omega}$ at several applied voltage amplitudes, starting from the amplitude of 50 V corresponding to the field just above E_c to 240 V. The increase of the voltage amplitude decreases the response time of the second harmonic signal only in the part after switching on the field. The response time after switching off the field does not change with the voltage amplitude. The response time τ was determined as a time necessary for $P_{2\omega}$ to change between 10% and 90% of the

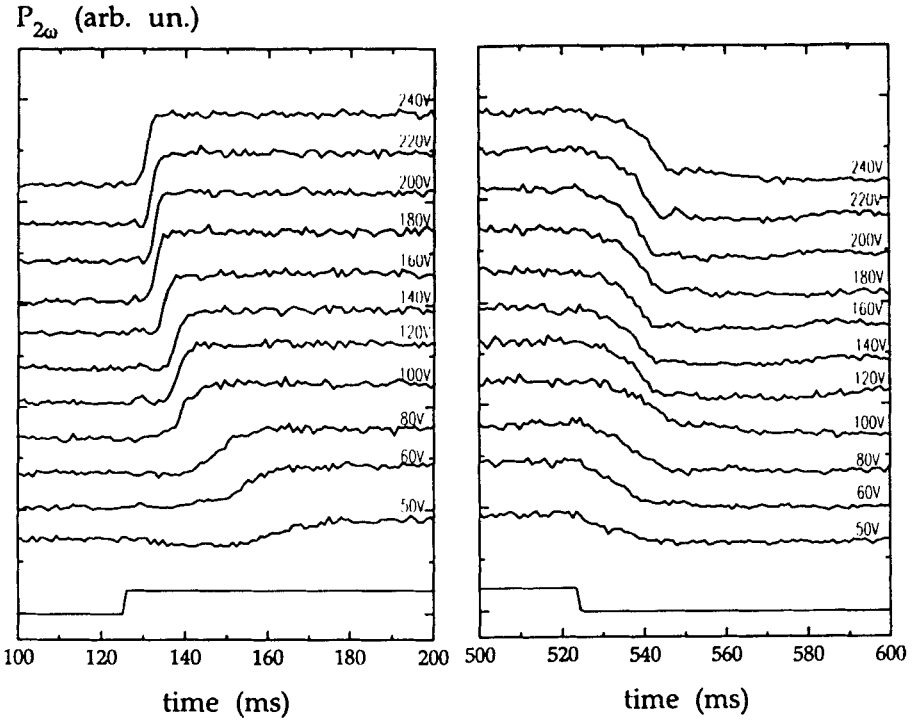

FIGURE 3 Temporal dependence of the SHG at different amplitudes of the modulating voltage at 2.5 Hz modulation. The voltage of 30 V corresponds to the critical field necessary to unwind the helical structure. Temporal dependence of the field is marked in the bottom of the figure.

maximal value. The explicit dependence of the inverse response times on the voltage amplitude is presented in Figure 4. The inverse response time after switching on the field $1/\tau_{on}$ increases linearly with the field. The inverse response time after switching off the field $1/\tau_{of}$ is constant. Its value of 50 Hz is very close to the relaxation rate of the

Goldstone mode of SCE9 determined by dielectric relaxation measurements [8]. These results imply that the dynamics of the winding and unwinding process, although including extreme changes of the SmC* helical structure, is mostly governed by a simple dynamic equation linear in applied field [9]

$$\gamma(\partial\varphi/\partial t) \sim P_s E \sin\varphi - K_3\theta^2(\partial^2\varphi/\partial z^2). \quad (1)$$

There φ is the phase angle of the director on the tilt cone, P_s is the spontaneous polarization, K_3 is the twist elastic constant, and γ is the viscosity of the material.

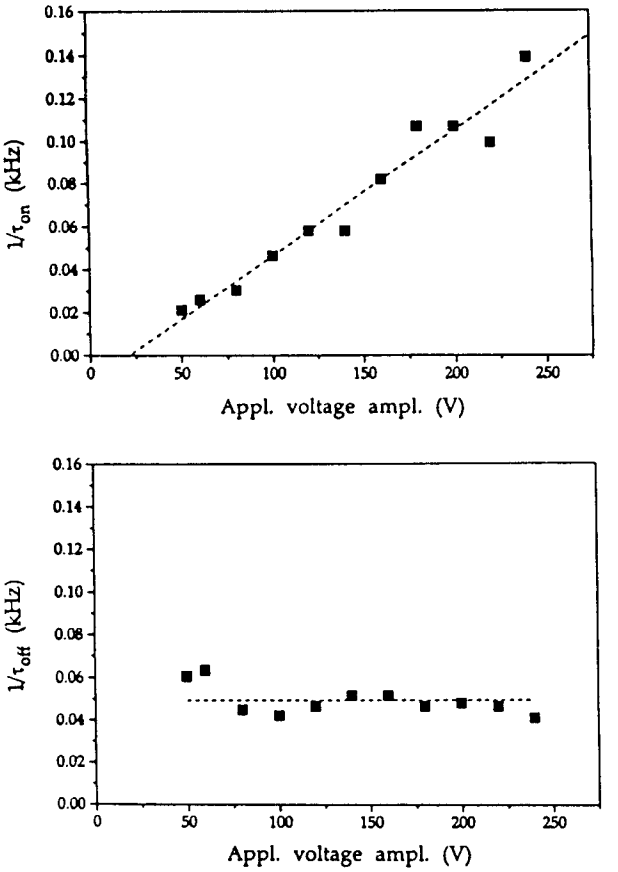

FIGURE 4 The dependence of the inverse response time of the SHG after switching the field on (τ_{on}) and after switching the field off (τ_{off}) on the amplitude of the applied voltage.

It was shown that at high applied voltage amplitudes the helical structure of the system unwinds within a few milliseconds. It is interesting to analyze how good is the molecular order established in such a short time interval and how does it differ from the structure present in the static external field. The answer is obtained by measuring the angular dependence of the modulated SHG. Figure 5 shows the temporal behaviour of $P_{2\omega}$ at different incident angles of the fundamental beam during the 2.5 Hz modulation with 220 V amplitude. The magnitude of $P_{2\omega}$ during the unwound state is changing almost identically to the characteristic phase matching curve observed in the DC field of the same magnitude (Fig. 6). This verifies that a complete homogeneous unwound structure is established in a few ms. On the other hand the absence of any transient $P_{2\omega}$ peaks at the angles different from the DC field phase matching angle of 2° shows that a strong coherent SHG can not appear until the structure is completely unwound.

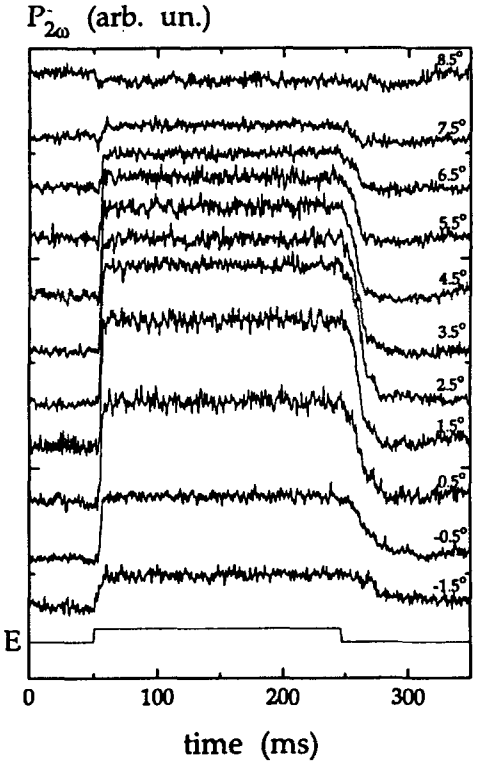

FIGURE 5 Temporal dependence of the SHG at different incident angles ϕ of the fundamental beam. The temporal dependence of the field is also marked.

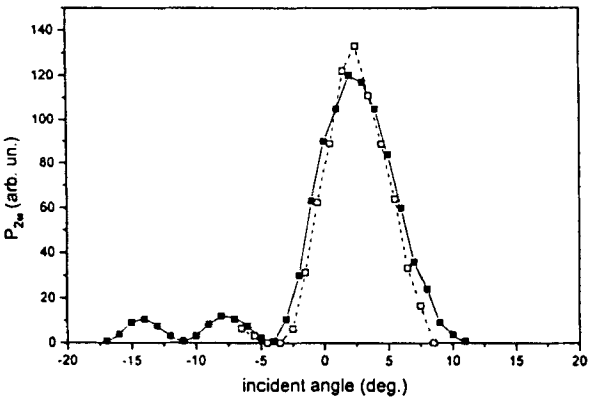

FIGURE 6 Angular phase matching curve in the DC field (filled squares) and in the 2.5 Hz square waveform field (open squares).

CONCLUSIONS

Our measurements present the possibility for effective on-off modulation of the optical SHG by ferroelectric liquid crystal in a thick homeotropic cell. The highest possible modulation frequency is determined by the spontaneous relaxation rate of the SmC* helical structure

$$1/\tau=(K_3/\gamma)(2\pi/p)^2 \quad (2)$$

which can be made higher by applying the materials with shorter helical pitch p and lower viscosity γ. Besides the possibility for modulation we have also shown that a study of SHG provides a lot of new information on the molecular dynamics during the complex switching process in the thick homeotropic FLC cell.

REFERENCES

1. J. Y. Liu, M. G. Robinson, K. M. Johnson, D. M. Walba, M. B. Ros, N. A. Clark, R. Shao, D. Doroski, J. Appl. Phys. **70**, 3426 (1991).
2. K. Schmitt, R. -P. Herr, M. Schadt, J. Funfschilling, R. Buchecker, X. H. Chen and C. Benecke, Liq. Crystals **14**, 1735 (1993).

3. D. S. Chemla, J. Zyss, Nonlinear Optical Properties of Organic Molecules and Crystals, (Academic Press, Orlando, 1987).
4. N. M. Shtykov, M. I. Barnik, L. A. Beresnev, L. M. Blinov, Mol. Cryst. Liq. Cryst. **124**, 379 (1985).
5. M. Utsumi, T. Gotou, K. Daido, M. Ozaki, K. Yoshino, Jpn. J. Appl. Phys. **30**, 2369 (1991).
6. M. Ozaki and K. Yoshino, Jpn. J. Appl. Phys. **28**, L 1830 (1989).
7. J. Y. Liu, M. G. Robinson, K. M. Johnson, D. Doroski, Opt. Lett. **15**, 267 (1990).
8. A. Levstik, Z. Kutnjak, C. Filipič, B. Žekš, Ferroelectrics **126**, [1087]/139 (1992).
9. B. Žekš, T. Carlsson, I. Muševič, B. Kutnjak-Urbanc, Liq. Crystals **15**, 103 (1993).