Appl. Phys. B 60, 485–487 (1995)

Applied Physics B Lasers and Optics

Helicoidal distributed-feedback cavity action in a ferroelectric liquid crystal

T. Furukawa, T. Yamada, K. Ishikawa, H. Takezoe, A. Fukuda

Department of Organic and Polymeric Materials, Tokyo Institute of Technology, O-okayama, Meguro-ku, Tokyo 152, Japan (Fax: + 81-3/5734-2876)

Received: 23 March 1994/Accepted: 4 October 1994

Abstract. Second-Harmonic (SH) generation was observed in homeotropically aligned ferroelectric liquid-crystal cells using fundamental light propagating along the helicoidal axis. Relatively strong SH light was observed in the helicoidal structure when the optical pitch was nearly the same as the SH-light wavelength, though, otherwise, the cancellation of the generated SH light by the helicoid results in negligibly weak SH intensity. Because the observed SH light is independent of the cell thickness, the SH light is attributed to the one generated from the surface region and the SH light generated inside the cell is confined and lost by the helicoid. This phenomenon indicates the helicoidal Distributed-FeedBack (DFB) cavity action and suggests the possibility of the helicoidal DFB laser using dye-doped ferroelectric liquid-crystal cells.

PACS: 42.80. Jr; 78.20. Wc

The Distributed-FeedBack (DFB) cavity is well known particularly in semiconductor lasers, where the feedback is provided by linear periodic modulations of the refractive index. Preiswerk et al. [1] succeeded in constructing a helical DFB gas laser using a hollow helical brass nut of pitch $L = 0.25$ mm. For the extension of the helical DFB laser to the visible-wavelength range, Kneubühl [2] proposed to use cholesteric liquid crystals doped with dyes. However, no one has realized the liquid-crystal helical DFB laser to date.

In order to confirm the helical DFB-cavity action in liquid crystals, optical higher-harmonic generation is very suitable, since a waveguide structure is not necessary; the generated light propagates along the same direction as the fundamental light because of the coherence phenomena. Recently, Kajikawa et al. [3] have already observed the helical DFB-cavity action in a ferroelectric smectic C* (SmC*) liquid crystal which has a helicoidal structure. They observed SH generation as a function of helicoidal pitch and found that not only the transmitted but also the reflected SH-light intensities markedly increase when the optical pitch of the helicoid coincides with the SH wavelength (Bragg condition); $\lambda_B = nP$, where n and P are the average refractive index and the nominal pitch of the helicoid, respectively. However, this increase is less than that attained by unwinding the helicoid by an electric field.

In this paper, we describe a ferroelectric liquid crystal which has a helicoid smoothly varying with temperature around the SH wavelength, so that fine structures, if any, can be detected in the temperature dependence of the SH-light intensity. The cell-thickness dependence of the maximum SH intensity was also measured to clarify the origin of the increase of the SH light at $P = \lambda_B/n$.

1 Experimental procedure

The sample used was a ferroelectric liquid crystal mixture (Hoffmann-La Roche 6304) which has a short helicoidal pitch in the ferroelectric SmC* phase of the following phase sequence:

Iso(64 °C) SmA(59 °C) SmC*(−14 °C) Cryst.

The material was sandwiched between two glass plates which were treated with homeotropic agent (polyimide, JSR-JALS-204), so that the helicoidal axis is parallel to the cell-surface normal. The cell thickness was controlled with PolyEthylene-Terephthalate (PET) spacers whose thicknesses were 25, 50 and 100 μm. These cells were placed in a copper oven in which the temperature was regulated with an accuracy of ± 0.02 °C.

The helicoidal pitch was assessed by observing the selective reflection peak due to the helicoidal structure. The reflection spectra were measured with a spectrometer equipped in a polarizing optical microscope (ORC, TFM-120AFT-PC).

The SHG measurements were performed using the fundamental beam of a Nd:YAG laser (Spectra Physics, DCR 11) as a light source. The pulse duration was 8 ns and the repetition frequency was 10 Hz. The output power was reduced to 1 mJ per pulse by Neutral Density (ND) filters. The linearly polarized fundamental light was impinging upon the cell under normal incidence. The SH light was detected through an IR-cut filter and an interference filter in the transmitted direction by a photomultiplier tube (Hamamatsu, R955). The signal was sent to a boxcar integrator (Stanford Research, SR 250) and was processed by a microcomputer (NEC, PC9801).

2 Results and discussion

Figure 1 shows the reflection spectra measured at several temperatures. All the peaks exhibit a simple symmetric shape and give the optical pitch *nP* of the helicoid. The reflectivity amounts to about 50%, so that one of the circularly polarized light beams with the same helical sense as that of the helicoid is almost completely reflected.

Figure 2 shows the temperature dependence of the SH-light intensity observed in 25, 50 and 100μm thick cells. The optical pitches determined by the reflection spectra are also shown in Fig. 2. The mutual SH intensities, though given in arbitrary units, can be compared with one another. The following characteristics can be noted: (i) The SH intensity shows an increase around 532 nm, the SH wavelength; (ii) The maximum SH intensities are almost independent of the cell thickness; (iii) The profiles of the SH-intensity variations are complicated and are different in the three cells.

The characteristic (i) clearly show the effect of the selective reflection on the generated SH light. At least the almost complete cancellation of the SH light due to the helicoid does not occur in the reflection regions, while it does occur in the other wavelength region. The characteristic (ii) suggests that the observed SH light originates from a certain thickness near the cell surface. The SH light generated inside the helicoid is seperated into two modes; one is a propagation mode and the other is a damping mode. The propagation mode is cancelled out due to the helicoid-like SHG in the other temperature range. The damping mode suffers the Bragg reflection back and forth throughout the cell, i.e., helicoidal distributed-feedback action, resulting in merely the loss of light intensity. Thus, only the two surface areas contribute to the emitted SHG. Recently [3], SH light was observed in both forward and backward directions. Notice that the damping mode in SmC* has such a polarization state that the electric field is parallel or perpendicular to the molecular long axis [4, 5], which is favorable for the constructive generation of SH light. Of course, the cell boundaries break this favorable condition because the optical eigenmodes are not orthogonal in the helicoidal medium [6].

In order to clarify the reason of the complicated temperature dependence of the SH intensity, i.e., characteristic (iii), and its reproducibility, we performed another SHG measurement using the same 25 μm cell. The optical pitch measurements were also carried out with minute temperature intervals. The results are shown in Fig. 3. It is easily noticed that the SH-intensity variation is reproducible; namely, the sudden increase occurs at the same temperatures. Very characteristically, the optical pitch shows jumps at the same temperatures when the SH intensity exhibits the sudden changes. Thus, the complicated SH-intensity variation with the several discontinuous changes is related to the stepwise changes of the optical pitch as a function of temperature. It is worth noting that the maximum SH intensity is obtained when the optical pitch is just 532 nm.

Why does the discontinuous change occur in the optical pitch? We can suggest the anchoring of the director

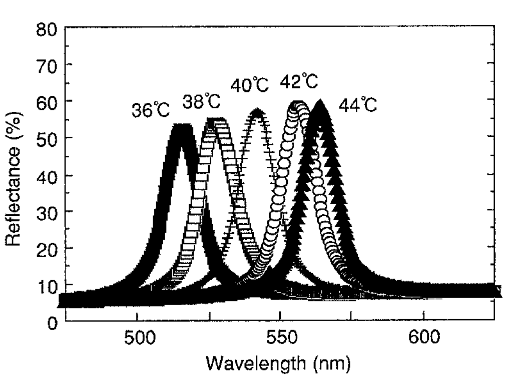

Fig. 1. Reflection spectra at several temperatures. The observed peaks are due to the selective reflection and are of simple symmetric shape

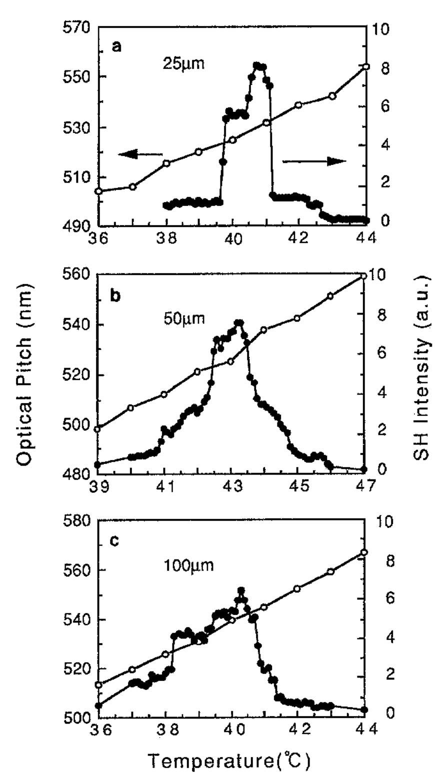

Fig. 2 a–c. Temperature dependence of the SH-light intensity and the optical pitch in three cells of different thickness; **a** 25 μm; **b** 50 μm; and **c** 100 μm. The increase of the SH-light intensity is observed when the optical pitch is about 532 nm, the SH-light wavelength

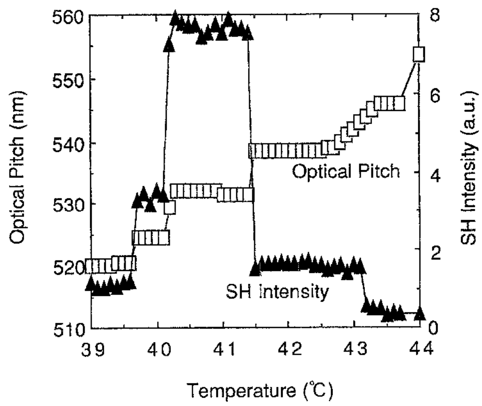

Fig. 3. Temperature dependence of the SH-light intensity and the optical pitch using the same 25 μm cell as in Fig. 1a. The pitch measurements with minute temperature intervals clearly indicate that the several discontinuous changes observed in the SH intensity and the optical pitch are mutually related. The stepwise change in the optical pitch originates from the surface-anchoring effect

on both glass surfaces. Since the nominal pitch P is 355 nm if we assume $n = 1.5$, there exist about 70 pitches in the 25 μm cell. Therefore, the increase or decrease of one pitch should give a discontinuous increase or decrease of the optical pitch by 1/70 of the pitch, i.e., about 7.5 nm, which is almost the same as the value obtained experimentally. The reason why the director is anchored even in the homeotropically aligned SmC* cell is not clear, but may be attributed to the process by which the material is introduced into the cell. The fact that the discontinuous changes in the SH intensity are much more pronounced in thinner cells supports the above interpretation.

Another interesting aspect is a prediction by Belyakov and Shipov [7, 8]. They showed theoretically that the enhancement of the nonlinear frequency transformation such as SHG occurs at the edges of the selective reflection band, and fine structure appears under the phase-matching condition. In the present case, the phase-matching condition is not realized. Actually, the SH intensity observed is not as strong as that from the unwound structure [3] so that the increase of the SH signal at around 532 nm is not an enhancement.

An enhancement can be expected when the dielectric constant of our system has non-zero imaginary part [2]. In this case, the SH light is possible to exhibit gain instead of loss. In this respect, the use of a dye-doped system will be a future problem. Other important future works are to reveal the polarization characteristics; how the phenomena are influenced by using right or left circularly polarized light, and what the polarization state of the emitted SH light is.

Acknowledgements. We thank Dr. M. Schadt of Hoffmann-La Roche, Switzerland, for supplying the liquid crystal used.

References

1. H.P. Preiswerk, G. Küttel, F.K. Kneubühl: Phys. Lett. A **93**, 15 (1982)
2. F.K. Kneubühl: Infrared Phys. **23**, 115 (1983)
3. K. Kajikawa, T. Isozaki, H. Takezoe, A. Fukuda: Jpn. J. Appl. Phys. **31**, L679 (1992)
4. Y. Ouchi, H. Takezoe, A. Fukuda, E. Kuze, N. Goto, M. Koga: Jpn. J. Appl. Phys. **23**, L464 (1984)
5. P. G. de Gennes, J. Prost: *The Physics of Liquid Crystals*, 2nd edn. (Clarendon, Oxford 1993) p. 276
6. A. Sugita, H. Takezoe, Y. Ouchi, A. Fukuda, E. Kuze, N. Goto: Jpn. J. Appl. Phys. **21**, 1543 (1982)
7. V.A. Belyakov, N.V. Shipov: Zh. Eksp. Teor. Fiz. **81**, 975 (1981)
8. V.A. Belyakov, N.V. Shipov: Phys. Lett. A **86**, 94 (1981)