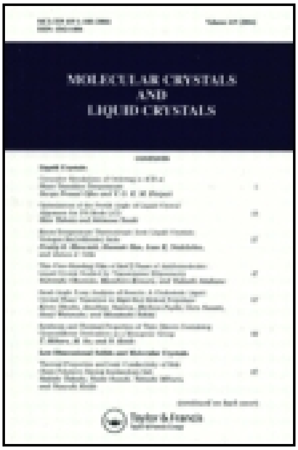

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Polarization Dependent Enhancement of Second-Harmonic Generation by Distributed Feedback Cavity Action in a Helicoidal Ferroelectric Liquid Crystal Cell

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A special phase matching for second harmonic (SH) generation was achieved in the helicoidal structure of a ferroelectric liquid crystal using normally incident two counter propagating waves, when the wavelength of the SH wave was near the selective reflection band edge. Theoretical prediction of the polarization dependence of the phase matched SH intensity was experimentally proved: (1) Two beams with the same circular polarization (e.g., R-R) give the most efficient enhancement. (2) When right- and left-circularly polarized light (R-L and L-R) beams are used, the SH light intensity is observed only in one side from which the circular polarization with the same handedness as that of the helicoid is incident, and is practically zero when observing in the opposite side. (3) The polarization of the enhanced SH wave is the same as the handedness of the helicoid of a ferroelectric liquid crystal regardless of the polarizations used for fundamental waves.

Keywords: second-harmonic generation; ferroelectric liquid crystal; smectic C* phase; helical structure; distributed feedback cavity; Bragg reflection; standing wave

INTRODUCTION

Phase matching of optical harmonic generation in the smectic-C* (Sm-C*) phase can be realized by a conventional angle phase matching by unwinding the helicoid by applying an electric field to homeotropic cells[1-3]. In addition to this phase matching, the helicoidal structure of the Sm-C* phase provides us with some particular possibilities to achieve an efficient harmonic generation. One of them was first exemplified by Shelton and Shen using Umkrapp process in a helical structure for the third-harmonic generation in a cholesteric liquid crystal (ChLC)[4-6]. Belyakov and Shipov[7-8] theoretically predicted that harmonic generation can be enhanced in ChLC when the harmonic wavelength is near the selective reflection band. Kajikawa *et al.*[9] and Furukawa *et al.*[10] succeeded in observing the second-harmonic generation (SHG) in the Sm-C* phase of ferroelectric liquid crystals using a single (unidirectional) fundamental wave when the wavelength of the SH wave matched with the optical pitch of a Sm-C* helix. Copic and Drevensek-Olenik[11-12] analyzed the effect theoretically, predicting that a special type of phase matching can be realized when two counter-propagating waves propagate along the helicoidal axis and generate a SH wave near the selective reflection band edge. Yoo *et al.*[13] showed the enhancement of SH light using two counter-propagating fundamental waves and confirmed the theory of Copic and Drevensek-Olenik. In the preceding paper, the dependence of the polarizations of fundamental waves on the enhancement were studied[14]. In this paper, more detailed experimental results on the polarization dependence are reported and are compared with theoretical results.

EXPERIMENTAL

The ferroelectric liquid crystal (FLC) mixture (ROLIC 6304) used in this study has a short helicoidal pitch covering the SH wavelength of a Nd:YAG laser (0.532 μm) in the ferroelectric Sm-C* phase with the following phase sequence: Iso (64°C) SmA (59°C) SmC* (-14°C) Cryst. It was homeotropically aligned between glass substrates, so that the helicoidal axis is normal to the substrate. The cell thickness was controlled by 75 μm thick polyethylene-terephthalate (PET) spacers. The real cell thickness was 87 μm which was measured by a fringe pattern of a vacant cell using a spectrometer (HITACHI U-3410).

The optical setup for the SHG measurements is illustrated in Fig. 1. A

Nd:YAG laser (Spectra Physics DCR-11) provided the fundamental pump wave (1.064 μm wavelength). We split the laser beam into two beams. After traveling about the same optical path length, two beams met each other at the sample from the opposite side with almost the same intensity. A He-Ne laser was used merely for optical alignment. The normal incident left or right polarizations of two counter propagating waves were produced by two quarter-wave plates, the direction of the optic axis of which was automatically controlled by a computer. The SH wave was detected by a photomultiplier (Hamamatsu R955) from the right-hand side of the sample after passing through a harmonic separator. The signal was sent to a boxcar integrator and analyzed by the computer.

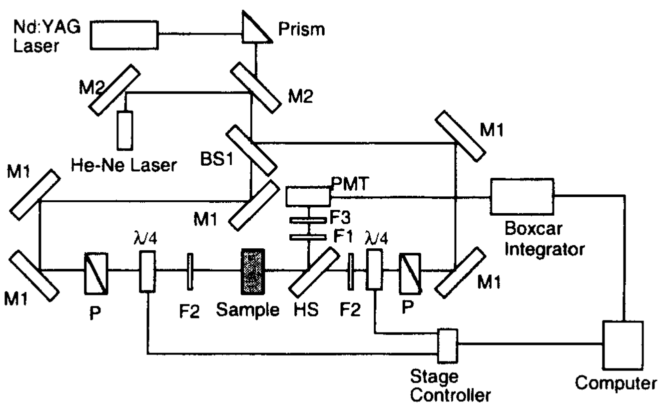

FIGURE 1 Optical arrangement for observing SHG. M: mirror, BS: beam splitter, HS: harmonic separator, F: filter, P: polarizer, λ/4: λ/4 plate, PMT: photomultiplier tube.

RESULTS AND DISCUSSION

The helicoidal pitch was determined by measuring transmittance spectra as a function of temperature. It smoothly increased with increasing temperature[10,13]. Figure 2 (a) shows the temperature dependence of the transmittance for the right-circularly polarized SHG of Nd:YAG laser (0.532

μm). The polarization was produced using a Fresnel Rhomb. It is clear in Fig. 2(a) that optical pitch matched with the SH wavelength at a sample temperature of about 42°C. The dip indicates that the ferroelectric liquid crystal has a right-handed helicoid.

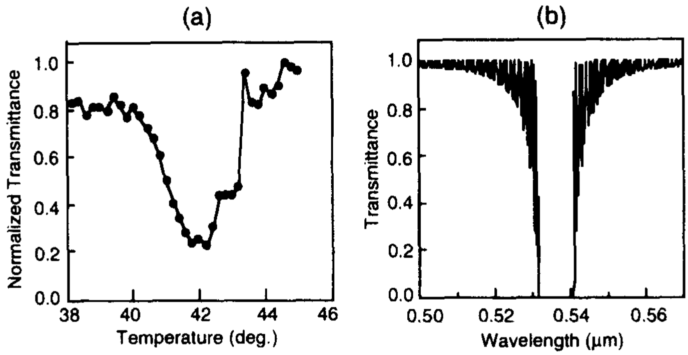

FIGURE 2. (a) Temperature dependence of a transmittance for a right-handed 0.532 μm wave. (b) Calculated selective reflection spectrum from a right-handed FLC helicoid for a right-handed circularly polarized wave.

The selective reflection spectrum for right-handed circularly polarized light propagating along the right handed helicoidal axis was simulated, as shown in Fig. 2(b). The following values were used for the simulation; ε_s (glass substrate) = 2.25, ε'_1 = 2.2, ε'_2 = 2.2 and ε'_3 = 2.8, which were measured by determining the deflection angle of an Ar laser by a wedge cell in the Sm-A phase. The tilt angle θ was 23° at 42°C, which was determined under a microscope by applying an AC electric field to a homogeneously aligned cell. Two edges of the selective reflection band are located at λ_2 and λ_{13}, which are expressed as

$$\lambda_2=\sqrt{\varepsilon'_2}\,p, \quad \lambda_{13}=\sqrt{\frac{\varepsilon'_1\varepsilon'_3}{\varepsilon'_1\sin^2\theta+\varepsilon'_3\cos^2\theta}}\,p,$$

where p is a pitch of a helicoid. The enhancement occurs when λ_2 matched with the wavelength of a SH wave[14]. Thus, $p = 0.35867$ μm was used for simulation, which correspond to $\lambda_2 = 0.532$ μm and $\lambda_{13} = 0.541$ μm.

Figure 3 (a) shows the temperature dependence of the SH light intensity under three polarization combinations for counter propagating fundamental waves. The pitch changes almost linearly with temperature in this range. R and L stand for the right- and left-circular polarizations of fundamental waves, respectively. The order of the first and second characters connected by a slash specifies the left and right incoming sides (Fig. 1) of the fundamental waves toward the sample. The detector was located in the right-hand side, as shown in Fig. 1. The enhancement was observed at both sides of the cell in the R-R combination because of the formation of a standing wave in the Sm-C* helicoidal structure. However, the SH light was generated only toward one side of the cell, if different polarizations such as R and L were used. It is also noted that the peak position under the L-R combination is slightly shifted from the one under the R-R combination. The temperatures, 42.6°C and 41.7°C, showing peaks for R-R and L-R combinations correspond to helicoidal pitches of 0.360 μm and 0.356 μm.

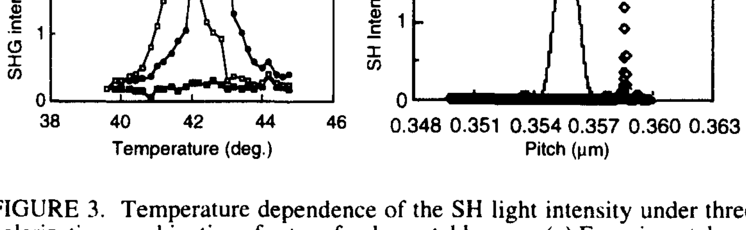

FIGURE 3. Temperature dependence of the SH light intensity under three polarization combinations for two fundamental beams. (a) Experimental and (b) theoretical results.

On the basis of the theory by Drevensek-Olenik and Copic[12], the polarization dependence of the SH wave intensity was simulated in Fig. 3 (b) when two counter propagating waves with different polarization combinations such as R-R, R-L and L-R propagate along the helicoidal axis of the cell. The calculation was made by taking account of the propagation of the fundamental and SH wave in the Sm-C* phase between two glass substrates. The following tentative values were used for $\chi^{(2)}$ components; $\chi_{123}=2$, $\chi_{112}=1$, $\chi_{332}=3$, $\chi_{222}=2$. The experimental results agreed well with the simulation except for the following points: (1) The experimental peak particularly under the R-R combination is broader than the theoretical one. This may be caused by the variation of the pitch caused by temperature fluctuation and surface anchoring. (2) The SH peak intensities for R-R and L-R combinations are almost the same, though about 25 times larger SHG is theoretically expected in R-R combination than in L-R combination. The reason of this large difference is not known at present. The lack of ideal circularly polarized light in the right-hand side because of the beam splitter inserted may be partly the reason.

The polarization of SHG was measured using the combination of a Fresnel Rhomb and a polarizer. Figure 4 shows the temperature dependence

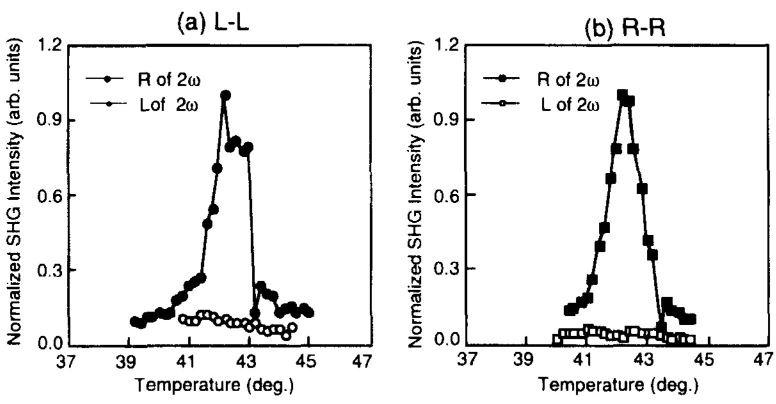

FIGURE 4. (a) Temperature dependence of R and L components of SHG for (a) L-L and (b) R-R fundamental waves.

of R and L components of SHG. Regardless of the polarizations of the fundamental waves, the component of polarization was right-handed circularly polarized. Hence, the enhanced SH light polarization is unambiguously the same as the handedness of the helicoid of the liquid crystal used, since the same circular polarization as the helicoid handedness is subjected to the Bragg reflection. Thus, it was experimentally confirmed that the distributed feedback cavity effect is caused by the Bragg reflection in the ferroelectric Sm-C* liquid crystal. The handedness of the circular polarization of the SH light was also confirmed to be the same as that of the helicoid theoretically.

CONCLUSION

We have studied the special phase matching in a helicoidal ferroelectric Sm-C* liquid crystal cell. Particular attention was paid to polarization conditions for the fundamental and SH waves. The same polarizations for the two counter propagating fundamental beams such as R-R generate the highest SH light. R-L or L-R combination gives a reasonable SHG, when the detection was made from the R side, if the helicoid is a right-handed one. Regardless of the polarization combinations for the fundamental waves, the handedness of the circular polarization of the SH wave was the same as that of the helicoid. It was confirmed that these experimental results agree with the theory by Drevensek-Olenik and Copic. For further simulation studies, it is necessary to determine the susceptibilities of the ferroelectric liquid crystal. This experiment is now progress in our laboratory. The effect of SHG-active-dye doping is also interesting and will be one of the future problems.

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