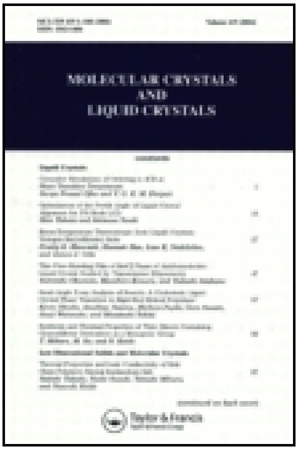

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Enhancement of Second-Harmonic Generation in Helicoidal Distributed Feedback Structure of a Ferroelectric Liquid Crystal Using Two Counter-Propagating Fundamental Waves

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Second-harmonic generation (SHG) was observed in an unwound ferroelectric smectic C* liquid crystal using two counter-propagating fundamental waves. A remarkable enhancement occurred when SH light wavelength approached to the optical pitch of the helicoid. This enhanced signal based on a helicoidal distributed feedback action is proportional to the fourth power of the sample thickness in thick cells, and becomes larger compared with the conventional phase-matched SHG signal in relatively thick samples. The crossover points of the SH intensities in the two phase matching types shift toward thinner cell thickness in materials with a larger dielectric anisotropy.

Keywords: second-harmonic generation, ferroelectric liquid crystal, helicoidal structure, distributed feedback cavity

INTRODUCTION

Shelton and Shen[1] first observed the inhomogeneous phase matching of the third-harmonic generation in cholesteric liquid crystals. Belyakov and Shipov[2] theoretically predicted that the harmonic generation can be enhanced in cholesteric liquid crystals when the harmonic frequency is near the selective reflection band. Actually, Kajikawa et al.[3] and Furukawa et al.[4] succeeded in observing SHG in FLCs, without unwinding the helicoid. Recently, Copic and Drevensek-Olenik[5] analyzed the effect theoretically, predicting that a special type of phase matching occurs when two counter-propagating fundamental waves propagate along the helicoidal axis and generate SH waves at the edge of the selective reflection band. In this paper, we experimentally confirmed the special type of phase matching using the suggested optical geometry. The results were compared with the theory of Copic and Drevensek-Okenik[5].

EXPERIMENTAL

The sample used was a ferroelectric liquid crystal mixture (ROLIC 6304) which has a short helicoidal pitch in the ferroelectric smectic C* phase. The material was introduced into a wedge-type cell to form a homeotropic alignment. The fundamental light of a Nd:YAG laser was used as a light source. The SHG measurements were performed using the two geometries for the fundamental waves, i. e., two counter propagating beams and a uni-directional beam[6]. For the experiments using two counter-propagating fundamental waves, we placed a mirror so that 1064-nm-wavelength light totally reflects back to the cell. The sample was set on a translational stage and the thickness dependence of SHG was observed by translating the wedge cell perpendicular to the optical path.

RESULTS AND DISCUSSIONS

At about 45°C, the optical pitch matches the SH wavelength, 532 nm, and smoothly increases with increasing temperature. Figure 1 shows the SH light intensity as a function of the optical pitch measured at three different positions of the wedge cell using two counter-propagating fundamental waves (open symbols). The sample thicknesses were about 35 μm, 50 μm and 75 μm. The result at the 75μm-thick position using a unidirectional fundamental wave is also

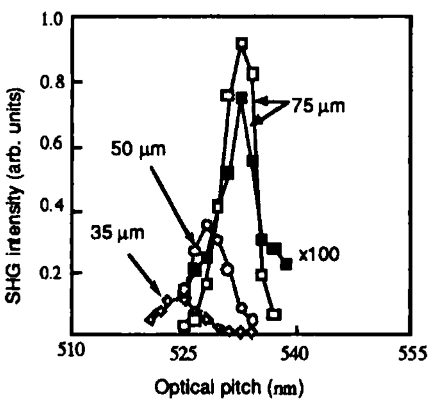

FIGURE 1. SHG intensities for 35 μm, 50 μm and 75 μm cell thicknesses. Open and closed symbols represent the results of the counter-wave and single-wave experiment, respectively. The scale for the single-wave experiment is enlarged by 100 times.

shown by closed symbols. The following features should be noticed: (1) the experiment using counter waves gives an SHG signal about 120 times larger than that of the experiment using a single wave; (2) the SHG signal increases with increasing cell thickness (the dependence is larger than quadratic); (3) the peak position shifts toward high optical pitch with increased cell thickness; and (4) the peak position does not differ between the experiments using counter and single waves.

The SHG intensity was simulated as a function of the cell thickness and the optical pitch based on the theory by Copic and Drevensek-Olenik[5]. The result shown in Fig. 2 well explains features (2) and (3) in Fig.1.

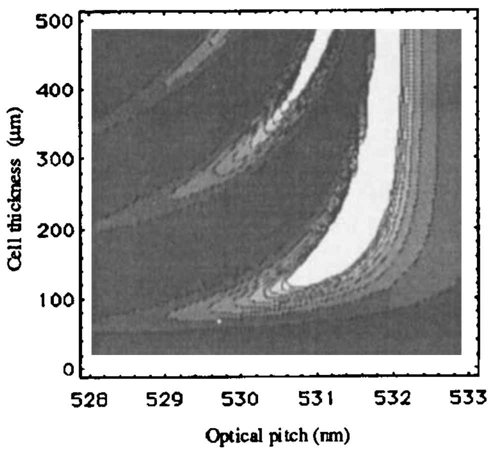

FIGURE 2. Simulated contour map of the SHG intensity. The brighter region represents higher SH intensity (n=1.5 and Δn=0.04).

Figure 3 shows the simulated SHG intensities in the conventional and special phase matching conditions. The parameters used for the calculation are the same as those in Fig. 2 except for using various Δn's. Note that the L^2 dependence is equivalent to the SH intensity change at 532 nm in Fig. 2, and that the $L^{2\sim4}$ dependence is equivalent to the plot along the ridge in Fig. 2. The crossover for the two phase matching mechanisms occurs in a thinner thickness when Δn becomes larger. Thus, we can obtain stronger SH light compared with that in the conventional phase matching using cells of the same thickness, if we use relatively thick cells of materials with a large birefringence.

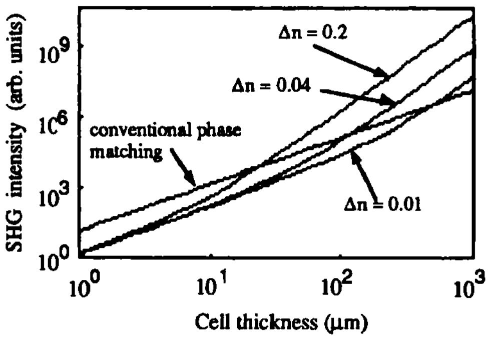

FIGURE 3. Simulated SHG intensity for the conventional and the special phase matching conditions (n = 1.5).

The higher L dependence of the SHG intensity than the quadratic dependence strongly indicates the presence of the special phase matching, namely, the helicoidal DFB (distributed feedback) cavity action. Copic and Drevensek-Olenik[5] suggested that the results of the previous experiments made by Kajikawa et al.[3] and Furukawa et al.[4] using a unidirectional fundamental wave are due to interaction of the incoming wave and a weaker wave that is reflected from the back surface of the sample. Features (1) and (4) in Fig. 1 support this Copic and Drevensek-Olenik's interpretation.

In conclusion, we confirmed a helicoidal DFB cavity action using two counter propagating fundamental waves along the helicoidal axis of a ferroelectric SmC* liquid crystal. The experiment qualitatively agrees with the theoretical prediction by Copic and Drevensek-Olenik[5].

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