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PERMITTIVITY GRADIENT INDUCED SECOND HARMONIC GENERATION IN PERIODIC NEMATIC STRUCTURE

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Abstract Nonlinear optical frequency doubling is studied in nematic liquid crystal cells where the director varies periodically in space. Flexoelectric polarization gives rise to a nonvanishing effective $\chi^{(2)}$; thus allowing bulk second harmonic generation.

INTRODUCTION

For reasons of symmetry, optical second harmonic generation (SHG) is forbidden in uniform bulk nematics in the dipole approximation. SHG may arise due to quadrupolar and higher order effects, but this contribution is expected to be small[1],[2]. In nonuniform nematics, the inversion symmetry is broken, and thus SHG is allowed. One mechanism for this is via the electric polarization arising from the spatial variation of the dielectric permittivity[3, 4].In nematics, this polarization is known as flexo- and order-electricity. The polarization gives rise to a D.C. electric field, and this field, together with the third-order nonlinear susceptibility, gives rise to an effective second-order susceptibility and the possibility of SHG. SHG has been observed in nematics due to director fluctuations[5].

In this paper, we report SHG from nematic cells with spatially periodic director variations. Using an optical buffing technique[6], nematic cells were constructed in which, due to spatial variations of the easy axis on the cell walls, the director varies as function of position within the cell. The resulting periodic flexoelectric polarization together with the third-order susceptibility give a spatially varying effective $\chi^{(2)}$. Using a mode-locked Nd:YAG laser, we have studied SHG from such periodic bulk samples to investigate this mechanism.

THEORETICAL BACKGROUND

The nonlinear wave equation in the nonuniform liquid crystal can be written as

$$\nabla \times \nabla \times \mathbf{E} - (\frac{\omega}{\mathbf{c}})^2 \varepsilon \cdot \mathbf{E} = (\frac{\omega}{\mathbf{c}})^2 \mathbf{P}_{\mathrm{NL}} \tag{1}$$

where the field $\mathbf{E}(\mathbf{r}, \mathbf{t})$ is time harmonic, and ε is the linear dielectric tensor. Because of the nonuniformity of the material, $\nabla(\nabla \cdot \mathbf{E})$ in the first term of Eq. 1 cannot be neglected. The nonlinear polarization $\mathbf{P}_{\mathrm{NL}}$, in the electric dipole approximation, can be written as

$$P_{NL,i} = \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l + ... \tag{2}$$

Even if $\chi^{(2)}_{ijk} = 0$, as in the case of a uniform nematic, second harmonic generation is possible in the presence of a d.c. field $\mathbf{E(0)}$ due to contributions to the nonlinear polarization such as

$$P_{NL,i}(2\omega) = \chi^{(3)}_{ijkl} E_j(0) E_k(\omega) E_l(\omega) \tag{3}$$

arising from Eq.2

Spatial variations of the nematic director give rise to DC electric polarization, and hence to a DC electric field. The simplest periodic director variation is

$$\hat{\mathbf{n}} = (\cos \frac{2\pi \mathbf{y}}{\mathbf{\Lambda}}, \sin \frac{2\pi \mathbf{y}}{\mathbf{\Lambda}}, \mathbf{0}) \tag{4}$$

where Λ is the wavelength of the director modulation. The dielectric tensor $\varepsilon(\mathbf{r})$ of the nematic liquid crystal is

$$\varepsilon(\mathbf{r}) = \varepsilon_{\perp} \mathbf{I} + (\varepsilon_{\parallel} - \varepsilon_{\perp}) \hat{\mathbf{n}}\hat{\mathbf{n}} \tag{5}$$

Since the flexoelectric polarization is proportional to the divergence of $\varepsilon(\mathbf{r})$ [3],[4], and if $\mathbf{E(0)} = \mathbf{a}\nabla \cdot \varepsilon(\mathbf{r})$, the effective $\chi^{(2)}$ induced by spatial variations of the dielectric permittivity is

$$\chi^{(2)}_{eff} = a\chi^{(3)} \cdot (\nabla \cdot \varepsilon(y)) \tag{6}$$

Because of the existence of periodic director modulation with wavevector $q = \frac{2\pi}{\Lambda}$, momentum conservation gives the phase matching condition for SHG as

$$\mathbf{k(2\omega)} = \mathbf{2k(\omega)} + \mathbf{2\mathit{l}q} \tag{7}$$

where $\mathbf{k}(\omega)$ is the wave vector of the fundamental in the medium, and l is an integer. A detailed theoretical description is given elsewhere[8].

SAMPLE PREPARATION

The nematic sample with periodic director modulation is shown in Fig. 1; the modulated structure was realized using optical buffing of the alignment layer on the cell walls.

The sample cell consists of two glass plates coated with an azo dye (Disperse Orange 3) doped polyimide alignment layer, separated by $40\mu m$ mylar spacers, and containing the nematic mixture E7.

Before filling the cell, the alignment layers were irradiated with polarized light from a CW Ar^+ laser. Due to alignment of the dye by the illuminating light, this gives rise to planar anchoring with the easy axis of the director at the surface perpendicular to the direction of polarization. By simultaneously translating the glass plate using a 2D translation stage and changing the direction of

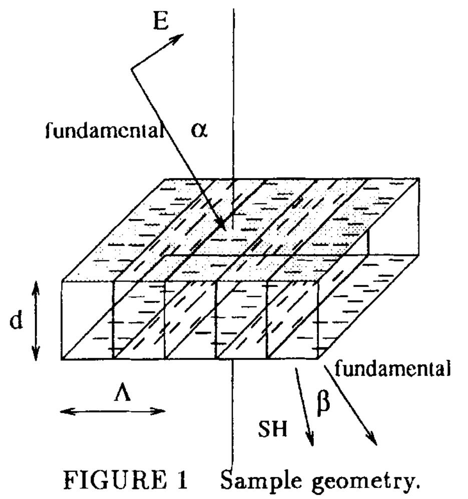

FIGURE 1 Sample geometry.

polarization of the write beam using an electrooptic modulator, high resolution anchoring patterns can be optically written on the alignment layers. Once the cell is filled, the nematic adopts the configuration which minimizes the free energy, and thus, by modulating the anchoring, modulated bulk structures may be realized[6]. We prepared substrates where the alignment direction was uniform in $25\mu m$ strips, but changed by 90^o between alternating strips. Cells were then capillary filled with E7 in the nematic phase. Preliminary results indicate that anchoring due to optical buffing is strong[7], we therefore expect that the director at the surface is essentially in the buffing direction at the surface except near the edges of the strips. Away from the surface, however, due to elastic forces, we expect the director modulations to relax with a characteristic length comparable to Λ.

EXPERIMENTAL SETUP AND RESULTS

A schematic of the experimental setup for second harmonic generation is shown in Fig. 2. A Nd:YAG laser with $38ps$ pulsewidth and $20mJ$ per pulse at $\lambda = 1.06\mu m$ was used as the source of radiation at the fundamental frequency. The energy incident on the sample was controlled by a $\lambda/2$-plate (WP) and polarizer (P); and was monitored by a fast photodetector (D) and oscilloscope. The polarization of the fundamental was along the wavevector of the director modulation in the sample; the

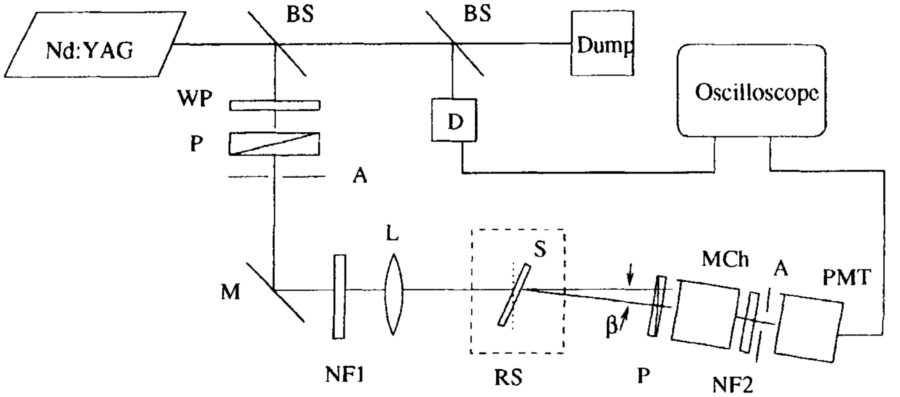

FIGURE 2 Experimental setup for second harmonic generation.

horizontal y-direction in our case. The sample (S) was mounted on a rotary stage (RS) with a vertical axis so that the angle of incidence could be varied. A notch filter (NF1) at 1.06 μm was situated in front of the sample and a monochromator (MCh) and a notch filter at 0.534 μm (NF2) were in front the photomultiplier (PMT), whose cutoff wavelength is 860 nm. With this setup, the typical signal to noise ratio in our experiment was in excess of 16dB.

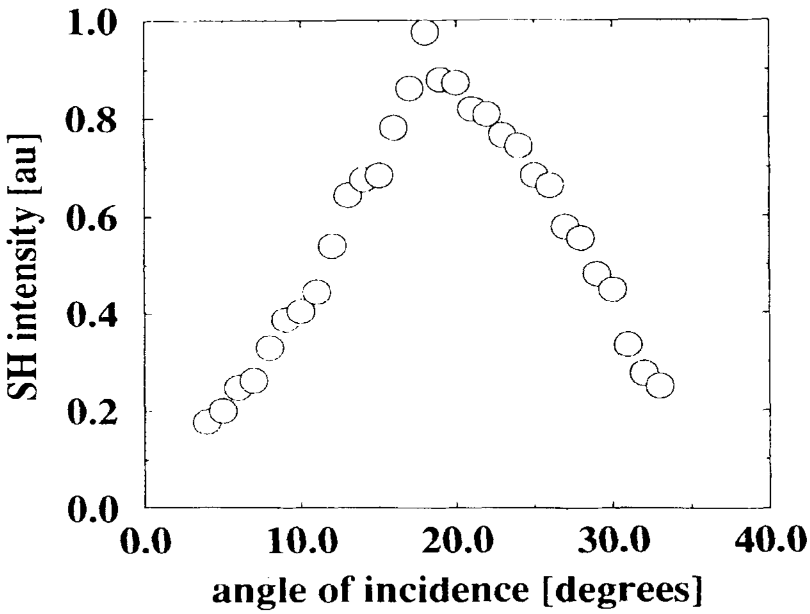

FIGURE 3 SH intensity vs. angle of incidence.

Strong second harmonic signal was observed from our samples when the angle of incidence of the fundamental was $\simeq 18^{\circ}$, as shown in Fig. 3.

Careful measurements further show that the angle between the outgoing fundamental wave and the SH wave is $\simeq 1.5^{\circ}$, in agreement with non-collinear phase matching suggested by Eq. 7. An approximate analytical calculation has been carried out to determine the phase matching conditions explicitly for a simple model; the details of this will be presented elsewhere[8]. Although a variety of phase-matching scenarios are possible, the results of calculations show that for E7 in our geometry, the optimum phase matching occurs when $l = -1$ in Eq.7, as shown in Fig.4.

The polarization state of SH signal is same as that of incident beam. The results of polarization measurements are shown in Fig. 5.

The second harmonic efficiency of our sample is approximately 3% of a $LiNbO_3$ crystal of the same length. From Eq.6, we expect the efficiency to scale as q^2; measurement of the dependence of second harmonic efficiency on the modulation wavevector is currently under way.

SUMMARY

We have observed SHG from nematic samples with periodic director distortions. The samples were constructed using optical buffing of photosensitive alignment layers. The nonlinearity responsible for SHG arises from the third-order susceptibility and the electric polarization arising from spatial variations of the dielectric permittivity. Optimum phase matching is found to be non-collinear, where the angle between the outgoing fundamental and the SH waves depends on the wavevector of the modulation.

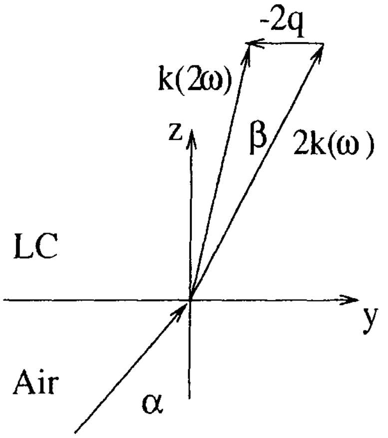

FIGURE 4 Phase matching condition.

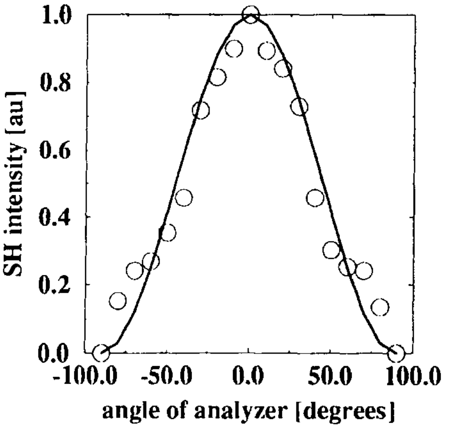

FIGURE 5 SH intensity vs. analyzer angle. The solid line corresponds to plane polarization along the y axis.

Since for this mechanism the conversion efficiency of periodic samples is expected to increase with the modulation wavevector, inhomogeneous liquid crystal structures with high spatial frequency may be efficient materials for SHG and other second order effects.

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