Enhancement of second harmonic generation in helical S_c liquid crystals

Martin Čopič[a]; Irena Drevenšek-Olenik[a]

[a] Department of Physics, Institute J. Stefan, University of Ljubljana, Ljubljana, Slovenia

PLEASE SCROLL DOWN FOR ARTICLE

Enhancement of second harmonic generation in helical S_C^* liquid crystals

by MARTIN ČOPIČ and IRENA DREVENŠEK-OLENIK*

Department of Physics and Institute J. Stefan, University of Ljubljana,
61000 Ljubljana, Slovenia

In a ferroelectric liquid crystal, a special type of phase-matching for optical second harmonic generation (SHG) is possible, where two counter-propagating fundamental waves create second harmonic waves at the edge of the selective reflection band. We compute the SHG intensity in such a situation and show that, at slight detuning from exact phase-matching, useful resonance enhancement can be obtained. A considerable amount of SHG also appears when the second harmonic frequency is in the reflection band, where the SHG wave is non-propagating.

1. Introduction

Optical second harmonic generation (SHG) in the ferroelectric S_C^* phase of liquid crystals is interesting both from the fundamental and applications points of view. The molecular hyperpolarizability of some materials forming the S_C^* phase is relatively large. If well oriented samples which are thick enough and which allow phase matching of the fundamental and second harmonic wave can be prepared, such systems are promising for efficient practical SHG devices.

In the equilibrium bulk phase, the S_C^* liquid crystal has a superstructure in which the molecular tilt and the spontaneous electrical polarization form a helix in the direction of the normal to the smectic layers. In most SHG experiments to date, the helix was unwound by the application of an external field [1–7]. In this case, the unwound S_C^* sample has a macroscopic C_2 symmetry and phase matching is achieved, as in ordinary crystals, with the use of birefringence.

The inhomogeneous helical structure, however, has some interesting optical properties which can be exploited for SHG phase matching. The dispersion relation for light propagating along the direction of the helix has two branches which are at most frequencies separated by approximately twice the wave-vector of the helix, 2q. In a small frequency interval, one of the branches has a gap in which the corresponding light wave cannot propagate, but is Bragg reflected on the helical superstructure [8]. This gives rise to the characteristic colouration of helical phases. The phase-matching condition for SHG can include a multiple of q

* Author for correspondence.

providing several new possibilities. The inhomogeneous phase-matching conditions for third harmonic generation in a chiral nematic liquid crystal have been analysed by Shelton and Shen [9–11] and for SHG in a twisted nematic under an external electric field by Saha [12].

2. Present work

In this paper we analyse a particularly interesting case of phase-matched SHG in the helical S_C^* phase where two fundamental waves propagate in opposite directions along the helical axis and the doubled frequency is near or in the reflection band. The induced polarization can generate the non-propagating wave in the gap. This phenomenon is very interesting, as this is a rare case where it is possible to study waves emitted by sources within the forbidden gap.

Experimentally, an enhancement of SHG in the vicinity of the gap has recently been observed by Kajikawa et al. [13] and Furukawa et al. [14]. They have suggested that the observed effect is analogous to the situation in distributed feedback laser resonators. That some enhancement of the harmonic generation could occur in a twisted nematic liquid crystal when the harmonic frequency is near the reflection band edge, has also been theoretically suggested by Belyakov [15, 16]. A satisfactory analysis of the effect, however, has been lacking.

The propagation of light along the helical axis (z-axis) in the S_C^* phase is best described on the basis of two circular polarizations. The eigenwaves have the form

$$\mathbf{E}_l = (a_l^+ \exp[i(k_l + q)z], a_l^- \exp[i(k_l - q)z])\exp(-i\omega t)$$
$$(1)$$

The mode index has the values $l = 1, \ldots, 4$. The $a_l^{\pm}$ are normalized components of the lth eigenwave polarizations and are not orthogonal. The wavenumbers k_l and frequency are connected by the dispersion relation

$$k_l = \pm \left[\left(\bar{\varepsilon} \frac{\omega^2}{c^2} + q^2 \right) \pm \left(4\bar{\varepsilon} \frac{\omega^2}{c^2} q^2 + \alpha^2 \frac{\omega^4}{c^4} \right)^{1/2} \right]^{1/2} \quad (2)$$

Here $\bar{\varepsilon}$ is the average value of the dielectric tensor for propagation in the z direction, and α is the dielectric anisotropy in the plane of the smectic layers:

$$\alpha = \frac{(\varepsilon_{33} - \varepsilon_{11})\varepsilon_{11} \sin^2 \theta}{2(\varepsilon_{11} \sin^2 \theta + \varepsilon_{33} \cos^2 \theta)} \quad (3)$$

θ is the tilt angle and ε_{ii} are the principal values of the approximately uniaxial dielectric tensor. The dispersion relation (2) is shown in figure 1.

Let us consider a slab of S_C^* material with thickness L and two counter-propagating waves along the z-axis at the fundamental frequency ω with wavenumbers $k_1^\omega = -k_2^\omega$ on the branches 1 and 2 in figure 1. When $\omega = \omega_+/2$ or $\omega_-/2$, we have the SHG wavenumber $k_{1,2}^{2\omega} = 0$. We have omitted the possible material dispersion of ε, which would cause the phase-matching to appear at ω slightly less than $\omega_\pm/2$. When $\omega_- < 2\omega < \omega_+$, the SHG wave falls in the non-propagating gap, where the wavenumber is imaginary, and it is a very interesting question what the resulting SHG signal is.

In order to compute the second harmonic wave which emanates from both surfaces of the S_C^* slab, it is necessary to solve the wave equation in the slab with non-linear polarization as the source term. The non-linear polarization contains terms with the spatial dependence given by $\exp(imqz)$, $m = \pm 1, \pm 3$. For SHG in the chosen case, only the terms with the first circular component given
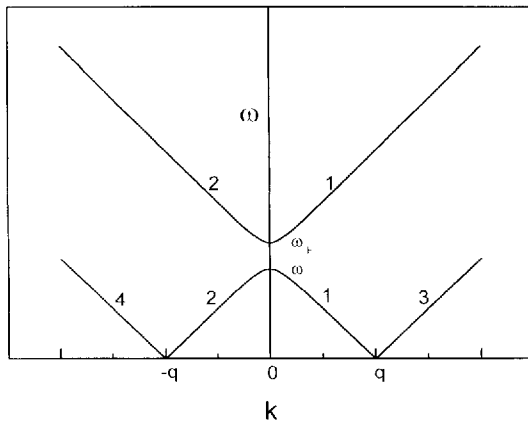
by $F^+ \exp(iqz)$ and the second one by $F^- \exp(-iqz)$ are effective. The amplitudes F^+ of the non-linear polarization at 2ω are determined by the amplitudes of the two fundamental waves and the non-linear susceptibility components of the S_C^* phase.

We will neglect the depletion of the fundamental beams due to the harmonic generation. The wave equation for the SHG wave is

$$\frac{d^2 E_{2\omega}^{\pm}}{dz^2} + \bar{\varepsilon} \frac{\omega^2}{c^2} E_{2\omega}^{\pm} + \alpha \frac{\omega^2}{c^2} E_{2\omega}^{\mp} \exp(\pm 2iqz) = F^{\pm} \exp(\pm iqz) \quad (4)$$

The usual approximation of the slow amplitude variation, reducing the second order equations to first order, is not applicable in the present case where $k_{1,2}^{2\omega} \simeq 0$. So the solution has to be sought in the form

$$E_{2\omega}^{\pm}(z) = \sum_{l=1}^{4} B_l a_l^{\pm} \exp\left[i(k_l^{2\omega} \pm q)z\right] + C^+ \exp(\pm iqz) \quad (5)$$

The first term is a sum of eigenwaves (1) and the last is a particular solution of equation (4) with the constants $C^{\pm}$ given by

$$C^{\pm} = \frac{F^{\pm}(\bar{\varepsilon}(\omega^2/c^2) - q^2) - F^{\mp} \alpha(\omega^2/c^2)}{(\bar{\varepsilon}(\omega^2/c^2) - q^2)^2 - \alpha^2(\omega^4/c^4)} \quad (6)$$

The denominator of the expression is zero for the frequencies $\omega_\pm$ at the band edge, so the coefficients are singular at least at one of the edges.

In the free space to the left and right of the slab we have two outgoing waves with amplitudes $D_{R,L}^+$, R, L standing for left and right side of the slab. We find them and the coefficients B_l by requiring that E and H are continuous at the boundaries. In general it is not possible to reduce the resulting system of 8 equations, because the eigenpolarizations of waves in the S_C^* phase are not orthogonal. However, of the four amplitudes B_l in equation (5), only the two belonging to branches 1 and 2 of the dispersion relation, for which $k_1^{2\omega} = -k_2^{2\omega} \simeq 0$ are nearly phase-matched, so they will give a dominant contribution to the SHG signal. Of the outgoing waves, only one circularly polarized wave on each side is significant. Its polarization is such that it would be Bragg reflected on normal incidence on the S_C^* slab, as would be expected. So we are left with only four equations for B_1, B_2, D_R^+ and D_L^-.

It is convenient to introduce two small quantities: $a = (\alpha/\bar{\varepsilon})^{1/2} = (\Delta n/n)^{1/2}$ and $t^2 = 1 - (k_1^{2\omega} + q)^2 c^2/(4\omega^2\bar{\varepsilon})$. In a typical S_C^* liquid crystal, a is close to 0·1 and with 2ω in the vicinity of the gap, t is of the same magnitude or smaller. The outgoing amplitude can be expressed to the

Figure 1. The dispersion relation for light waves propagating along the z axis in a twisted S_C^* liquid crystal.

order a^4 and t^4 in the form

$$D_R^+ = -\mathrm{i}\sin\frac{k_1^{2\omega}L}{2}$$

$$\times\left[\frac{a^2 - t^2}{a^2\exp(-\mathrm{i}k_1^{2\omega}L/2) + t^2\exp(\mathrm{i}k_1^{2\omega}L/2)}(C^+ - C^-)\right.$$

$$\left.+\frac{a^2 + t^2}{a^2\exp(-\mathrm{i}k_1^{2\omega}L/2) - t^2\exp(\mathrm{i}k_1^{2\omega}L/2)}(C^+ + C^-)\right]$$

$$(7)$$

The expression for D_L^- is similar. The first term in the square bracket gives a peak SHG amplitude at the upper edge of the reflection band ω_+ and the second at the lower edge ω_-.

Let us take $C^- = -C^+$. This will hold if $F^- = -F^+$, a condition that can be met with a proper choice of the polarizations of the two waves at the fundamental frequency. Then we get only one peak around $2\omega = \omega_+$. Let us write $2\omega = \omega_+(1 + r)$. r measures the amount of detuning from the perfectly phase-matched condition $k_1^{2\omega} = 0$. In terms of a and r,

$$k_1^{2\omega} \simeq \frac{\omega}{c}[\bar\varepsilon r(a^2 + r)]^{1/2} \qquad (8)$$

Taking into account equations (6) and (7), the SHG intensity emitted to the right is given, for positive r, approximately by

$$I^{2\omega}(L) = \frac{c^2|F^+|^2}{8\bar\varepsilon\omega^2 r^2}\frac{(a^2 - t^2)^2(1 - \cos k_1^{2\omega}L)}{a^4 + t^4 + 2a^2t^2\cos k_1^{2\omega}L} \qquad (9)$$

$I^{2\omega}(L)$, shown in figure 2, is a periodic function corresponding to the Maker fringes in the case of homogeneous crystals. It has, however, some interesting properties. At
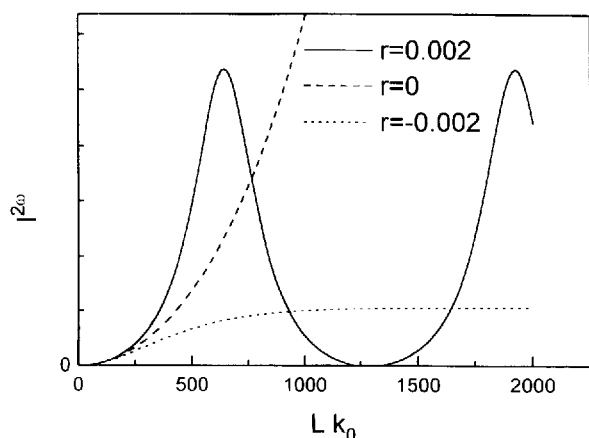

Figure 2. The dependence of the SHG intensity on thickness for three values of the detuning of second harmonic frequency from the edge of the selective reflection band. $a = 0.1$.

the maxima, which occur when $k_1^{2\omega}L = (2N + 1)\pi$, where $N = 0, 1, \ldots$, its value is $c^2|F^+|^2/(4\bar\varepsilon\omega^2 r^2)$. As shown in figure 2, for small detuning r, this value can be considerably larger than in the perfectly phase-matched case with the same L, where we have $I_{r=0}^{2\omega}(L) = (1/4)L^2|F^+|^2$ and the ratio of the first peak of equation (9) and $I_{r=0}^{2\omega}$ at the same L is

$$\frac{I^{2\omega}(L_{\max})}{I_{r=0}^{2\omega}(L_{\max})} = \frac{4(a^2 + r)}{\pi^2 r} \qquad (10)$$

Compare this to the usual case of a homogeneous crystal, where the first maximum of the Maker fringes is always smaller than the phase matched SHG intensity at the same wavelength by a factor $4/\pi^2$. The enhancement of $I^{2\omega}$ is similar to light amplification in distributed feedback lasers, although it occurs at small detuning from the reflection band edge on the side where light waves can propagate.

In a real material, a can be about 0.1. In the maximum, r and $L_{\max}$ are connected through $k_1^{2\omega}L_{\max} = \pi$ and equation (8). If we take L to be 600 pitch lengths or about 0.3 mm for green doubled light, which is a somewhat optimistic value for a well oriented sample, we get $r = 0.002$. Then the enhancement ratio (10) is 2.4, a value that could be quite useful.

When 2ω is in the reflection band, that is when $r < 0$, $k_1^{2\omega}$ is imaginary and the double light cannot propagate in the S_C^* liquid crystal. So it is an interesting question what happens to $I^{2\omega}$. Again for the case $F^- = -F^+$, we can derive from equation (7) a simple approximate expression

$$I^{2\omega}(L) = \frac{c^2|F^+|^2}{4\bar\varepsilon\omega^2|r|}\frac{\cosh\kappa L - 1}{a^2(\cosh\kappa L + 1) + 2r} \qquad (11)$$

Here $\kappa = -\mathrm{i}k_1^{2\omega}$. This expression is also shown in figure 2. The SHG intensity in the gap saturates at a value which one would get approximately in a phase-matched situation with the material thickness $1/\kappa$, as would be expected.

In a sample of S_C^* material, it is often more convenient to scan the pitch vector $\mathbf{q}$ by changing the temperature than to vary the sample thickness. In our notation, this is equivalent to changing the detuning parameter r. Figure 3 shows the dependence of $I^{2\omega}(L)$ at fixed L as a function of r at several choices of the components of the non-linear polarization $F^\pm$. With $F^- = -F^+$ we get a peak at the upper gap edge ω_+, with $F^- = F^+$ a peak at ω_-, and with $F^- = 0$ (or $F^+ = 0$) two peaks at both edges, with somewhat larger intensity in the gap, a consequence of the interference of the two terms in equation (7).

The peak in SHG intensity obtained by temperature tuning $\mathbf{q}$ through the reflection band has already been
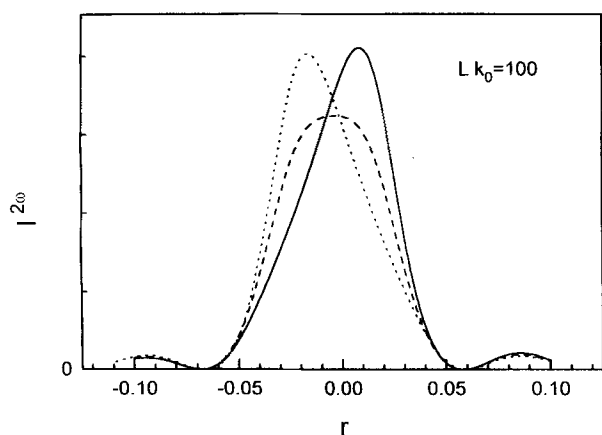
Figure 3. The dependence of the SHG intensity on the detuning parameter at fixed thickness of the sample. The three curves are for different choices of the components of the non-linear polarization: full line—$F^+ = -F^-$, dotted line—$F^+ = F^-$, dashed line—$F^- = 0$. $a = 0.1$.

observed by Kajikawa *et al.* [13] and Furukawa *et al.* [14]. Unfortunately, the authors did not use two counter-propagating fundamental waves, so their result is probably due to interaction of the incoming wave and a weaker wave that is reflected from the back surface of the sample.

For applications, materials with large optical anisotropy and large tilt, and a correspondingly larger value of the parameter a, might be found. A value of $a = 0.2$ seems quite possible, giving an enhancement ratio of 10 for a 300 μm thick sample. It is also much easier to prepare relatively thick samples with homeotropically twisted geometry, than with a planar geometry, with an electric field to unwind the helix. Well oriented homeotropic samples of a few hundred microns thickness can

usually be prepared without much trouble, whereas in the planar geometry problems start at 10 μm. Together with the high hyperpolarizability coefficients that can be obtained with organic materials, S_C^* liquid crystals could be very efficient systems for practical SHG applications.

References

[1] SHTYKOV, N. M., BARNIK, M. I., BERESNEV, L. A., and BLINOV, L. M., 1985, *Mol. Cryst. liq. Cryst.*, **124**, 379.

[2] TAGUCHI, A., OUCHI, Y., TAKEZOE, H., and FUKUDA, A., 1989, *Jpn. J. appl. Phys.*, **28**, L997.

[3] OZAKI, M., and YOSHINO, K., 1989, *Jpn. J. appl. Phys.*, **28**, L1830.

[4] LIU, J. Y., ROBINSON, M. G., JOHNSON, K. M., and DOROSKI, M., 1990, *Opt. Lett.*, **15**, 267.

[5] OZAKI, M., UTSUMI, M., GOTOU, T., DAIDO, K., and YOSHINO, K., 1991, *Jpn. J. appl. Phys.*, **30**, L1569.

[6] UTSUMI, M., GOTOU, T., DAIDO, K., OZAKI, M., and YOSHINO, K., 1989, *Jpn. J. appl. Phys.*, **30**, 2369.

[7] OZAKI, M., UTSUMI, M., GOTOU, T, MORITA, Y., DAIDO, K., SADOHARA, Y., and YOSHINO, K., 1991, *Ferroelectrics*, **121**, 259.

[8] DE VRIES, H. P., 1951, *Acta Crystallogr.*, **4**, 219.

[9] SHELTON, J. W., and SHEN, Y. R., 1970, *Phys. Rev. Lett.*, **25**, 23.

[10] SHELTON, J. W., and SHEN, Y. R., 1971, *Phys. Rev. Lett.*, **26**, 538.

[11] SHELTON, J. W., and SHEN, Y. R., 1972, *Phys. Rev. A*, **5**, 1867.

[12] SAHA, S. K., 1981, *Opt. Commun.*, **37**, 373.

[13] KAJIKAWA, K., ISOZAKI, T., TAKEZOE, H., and FUKUDA, A., 1992, *Jpn. J. appl. Phys.*, **31**, L679.

[14] FURUKAWA, T., YAMADA, T., ISHIKAWA, K., TAKEZOE, H., and FUKUDA, A, (to be published).

[15] BELYAKOV, V. A., and SHIPOV, N. V., 1981, *Phys. Lett.*, **86A**, 94.

[16] BELYAKOV, V. A., and SHIPOV, N. V., 1982, *Sov. Phys. JETP*, **55**, 674.