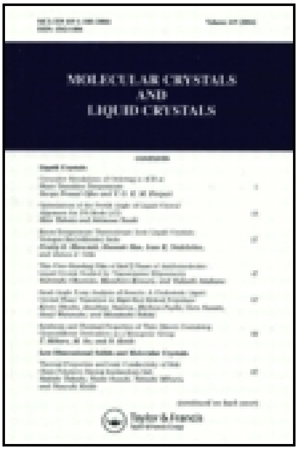

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

NONLINEAR SECOND HARMONIC GENERATION IN CHIRAL SMECTICS FOR FUNDAMENTAL WAVE BEING AT THE EDGE OF SELECTIVE REFLECTION BAND

V. A. Belyakov [a]

[a] L.D. Landau Institute for Theoretical Physics, Kosygin str. 2, 117334, Moscow, Russia

Version of record first published: 24 Sep 2006

Mol. Cryst. and Liq. Cryst., 2001, Vol. 366, pp. 491-504
Reprints available directly from the publisher
Photocopying permitted by license only

Nonlinear Second Harmonic Generation in Chiral Smectics for Fundamental Wave Being at the Edge of Selective Reflection Band

V.A. BELYAKOV

L.D. Landau Institute for Theoretical Physics, Kosygin str. 2, 117334 Moscow, Russia

It was theoretically predicted [1–4] and confirmed experimentally [5–7] that an enhancement of the second nonlinear optical harmonic generation (SHG) in ferroelectric smectics C* (Sm-C*) liquid crystals (LC) occurs if the SH frequency is close to the edge of selective reflection band for Sm-C*. The SHG enhancement (SHGE) occurs also when the both fundamental and SHG frequencies are simultaneously close to the edges of selective reflection bands in Sm-C* for first and second diffraction orders, respectively [8]. Because the enhancement of nonlinear frequency transformation is not specific only of Sm-C* and may be of great applied importance [8–10] results of theoretical investigation of the role of diffraction of fundamental wave in the SHG enhancement are presented. For SHG in Sm-C* for collinear geometry with the waves propagation direction along the helix axis behaviour of the fundamental wave and nonlinear polarizations are investigated at fundamental frequency close to the edge of second order reflection band [11]. It is found that all terms of nonlinear polarization are sharply increased at approach of the fundamental frequency to the edge of selective reflection band. The analytical investigation of the problem is illustrated by calculations in the framework of dynamical diffraction theory [11].

Keywords: second harmonic generation; light diffraction; Sm-C*

INTRODUCTION

Recently an enhancement of the second nonlinear optical harmonic generation (SHG) in ferroelectric smectics C* (Sm-C*) liquid crystals (LC) (see also the theoretical works [1-4]) was observed [5-7]. The SHG enhancement took place when the SHG frequency coincides with the edge of selective reflection band in Sm-C* and was especially strong if the pumping wave was a standing wave obtained by two counter propagating plane fundamental waves. General explanation of the observation was attributed to the enhancement of nonlinear frequency conversion effect predicted early theoretically for nonlinear periodic media [1-4, 9-11]. Complete interpretation of the observed SHG enhancement was presented in the papers [12, 13]. Under the experimentally studied conditions [5-7] the phase-matching conditions (PM) for SHG were independent on the frequency dispersion of the linear dielectric susceptibility. As the theoretical works show [1-4, 8, 12-15] in the general case the SHG enhancement occurs if some relations between the frequency dispersion of the linear dielectric susceptibility and other dielectric parameters are fulfilled. However for special configurations of the pumping field, for example the pumping wave as a standing wave [7,16], there is no need in the mentioned relations. Recently it was also shown that a dispersion independent enhanced phase-matched SHG may be also reached if the both fundamental and SHG waves are diffracting in a sample [8]. It is why a further investigation of the SHG enhancement and of the role of the fundamental wave diffraction, in particular for frequency dispersion independent SHG enhancement, is urgent. The analysis performed below shows that an enhancement of nonlinear polarizations in the case of the fundamental wave diffraction contributes to the SHG enhancement and reveals the conditions which are favourable for the polarizations and SHG enhancement.

PRINCIPAL EQUATIONS

Examine SHG in a planar Sm-C* layer assuming that the fundamental wave propagating along helical axes is subjected to diffraction in second diffraction order which alone is allowed for a wave propagating along the helix axes and corresponds to a circular polarization of the wave. It

means that the SHG wave is far enough from diffraction conditions and may be regarded almost as a plane wave.
To describe SHG one has to solve the following equation

$$\nabla\times\nabla\times E(r,2\omega) - (2\omega/c)^2\varepsilon E(r,2\omega) = (2\omega /c)^2\chi : E(r,\omega) E(r,\omega) \qquad (1)$$

where χ and ε are the quadratic nonlinear susceptibility and dielectric susceptibility of the Sm-C* at the frequency 2ω respectively and E(r,2ω), E(r,ω) are the fields of the second harmonic and fundamental wave. The solution should also satisfy boundary conditions. The form of the dielectric susceptibility of Sm-C* is discussed in details in the references [11, 13, 17].

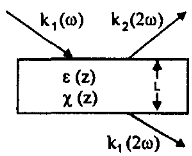

FIGURE 1 Schematics of SHG in Sm-C* at diffraction of the fundamental wave.

We apply for solution of the problem the two-wave approximation of the dynamical diffraction theory [11,17]. Being presented inside the sample as a superposition of two plane waves with the amplitudes E_1 and E_2 the fundamental field satisfies the following equations

$$\begin{aligned}(1 - (k_1/\kappa_\sigma)^2) E_1 + \delta_1 E_2 &= 0\\ \delta_1 E_1 + (1 - (k_2/\kappa_\sigma)^2) E_2 &= 0\end{aligned} \qquad (2)$$

The two eigen solutions of the system (2) are of the form

$$E(r,\omega) = (E_1 \exp[ik_1 r] + E_2 \exp[ik_2 r]) \exp[-i\omega t]), \qquad (3)$$

where $k_2 - k_1 = 2\tau$, and τ is the reciprocal vector of-the Sm-C* periodical structure. The field of fundamental wave is presented in the sample by a superposion of two eigen waves with the weights determined by the

sample thickness. We shall distinguish below these eigen waves and mark related to them quantities by a subscript ±.
In the general case the SH wave is presented in the sample by a linear superposition of two eigen solutions (almost plane waves) propagating in the opposite directions. So the field for the frequency 2ω is of the form $E(r,2\omega) = (E_1 \exp[ik_1 r] + E_2 \exp[ik_2 r]) \exp[-i2\omega t])$, where $k_2 - k_1 = 2\tau$.
Note that in our case of the SH being far enough from the diffraction conditions one of the amplitudes (E_1 or E_2) is much less than the other.
In the nondepleted pump approximation one gets from Eq.(1) the following nonhomogeneous linear equations for $E(2\omega)_1$ and $E(2\omega)_2$.

$$\begin{aligned} (1-(k_1/\kappa)^2)E_1 + \delta_2 E_2 &= -(4\pi/\varepsilon)P_{-n\tau}\delta(k_1 + n\tau - k_1(\omega)_\pi - k_2(\omega)_\sigma) \\ \delta_2 E_1 + (1-(k_2/\kappa)^2)E_2 &= -(4\pi/\varepsilon)P_{n\tau}\delta(k_2 - n\tau - k_1(\omega)_\pi - k_2(\omega)_\sigma) \end{aligned} \tag{4}$$

where κ is the modulus of the wave vector for the frequency 2ω, i.e. $\kappa^2 = \varepsilon(2\omega)(2\omega/c)^2$, $P_{-\tau}$ and P_τ are the nonlinear polarizations corresponding to the relevant Fourier harmonics in the expansion of the nonlinear quadratic nonlinear susceptibility χ, n=0,1,2, and $k(\omega)_\pi$ and $k(\omega)_\sigma$ are wave vectors of the fundamental wave provided by polarization indices [11,17].
The phase-matching conditions follow directly from the Eq.(4) and are determined by zero values of the arguments of δ-functions in (4).

$$k(2\omega)_1 + n\tau - k(\omega)_{1\pi} - k(\omega)_{2\sigma} = 0. \tag{5}$$

To obtain an explicit form of the phase-matching conditions (PM) (5) one has to know solutions for the eigen modes of the linear optics at frequencies ω and 2ω relevant to the wave vectors entering Eq.(4).

NONLINEAR POLARIZATIONS

Under diffraction of the fundamental wave the nonlinear polarizations $P_{-n\tau}$, $P_{n\tau}$ entering the right hand side of Eq.(4) have very unusual dependence on the sample thickness and the deviation of the pumping wave from the exact Bragg condition. One easily reveal this dependence taking into account that the coefficients in the linear combination of two eigen solutions (3) representing the fundamental field in a sample are dependent on the thickness and the deviation from Bragg condition. Because the nonlinear polarization in Eq.(1) is biquadratic in the amplitudes of plane waves constituting the fundamental field in the sample (a linear superposition of the eigen modes) the right hand sides of the system (4) contains terms which are proportional to the all possible products of coefficients determining the weight of these eigen solutions in the mentioned superposition:

$$C_+ = [E_0 \xi_{1-} \exp(-i\alpha_{1-} l/2)] / (\xi_{1-} \exp(-i\alpha_{1-} l/2) - \xi_{1+} \exp(i\alpha_{1+} l/2))$$
$$C_- = [E_0 \xi_{1+} \exp(i\alpha_{1+} l/2)] / (\xi_{1-} \exp(-i\alpha_{1-} l/2) - \xi_{1+} \exp(i\alpha_{1+} l/2)), \quad (6)$$

where E_0 is the pumping wave amplitude out of the sample, $l = \tau L$ is the dimensionless sample thickness, $\xi = E_2/E_1$ is the ratio of plane wave amplitude in the eigen solution, $\alpha_{1\pm}$ is determined by Eq. (9) (see below), the subscript + marks the eigen solution attenuating into the depth of the sample, the subscript - marks the eigen solution growing into the depth of the sample. In the limit of infinite sample thickness $C_+ = E_0$ and $C_- = 0$. It follows from Eqs.(6) that three different terms of the nonlinear polarization P_{++}, P_{+-} and P_{--} are entering in the right hand sides of the system (4):

$$P_{++} \propto E^2_0 \xi_{1-} \{[E_0 \exp(i\alpha_{1-} l/2)] / (\xi_{1-} \exp(-i\alpha_{1-} l/2) - \xi_{1+} \exp(i\alpha_{1+} l/2))\}^2$$
$$P_{--} \propto E^2_0 \xi_{1+} \{[E_0 \exp(i\alpha_{1+} l/2)] / (\xi_{1-} \exp(-i\alpha_{1-} l/2) - \xi_{1+} \exp(i\alpha_{1+} l/2))\}^2 \quad (7)$$
$$P_{+-} \propto -E^2_0 [\exp((i\alpha_{1+} + i\alpha_{1-}) l/2)] / \{(\xi_{1-} \exp(-i\alpha_{1-} l/2) - \xi_{1+} \exp(i\alpha_{1+} l/2))\}^2$$

All three expressions in (7) reveal sharp maxima at the edges of reflection band (RB) of the pumping wave as functions of the deviation from the Bragg condition (changing of the helical pitch or the pumping frequency). Out of the RB P_{++}, P_{+-} and P_{--} are dying out as functions of the frequency deviation from RB. Inside RB P_{++}, P_{+-} and P_{--} are dying out also as functions of the frequency deviation from the RB edge. The discussed dependence of P_{++}, P_{+-} and P_{--} on the sample thickness predetermines also an unusual dependence of SGH on the sample thickness. For the nonlinear polarizations P_{++}, P_{--} and P_{+-} the PM condition can be reached close to the RB edge for the terms of nonlinear susceptibility χ_0 and $\chi_{2\tau}$, respectively.

PHASE-MATCHING CONDITIONS

To write down an explicit form of the PM conditions (5) one have to find solutions of Eqs.(2) and (4). For light propagating along the helix there is only the second order diffraction in Sm-C* and, as was already mentioned, a polarization separation occurs [11,17] resulting in the circular eigen polarizations only one of which is subjected to diffraction. To be definite it will be assumed below that the polarization of the fundamental wave is diffracting circular one. Introducing a conventional parametrization of the both solutions for ω and 2ω frequencies one finds, for example, for the fundamental frequency the solution in the following way. The mentioned parametrization is of the form

$$k1n(\omega) = -\tau(1+\alpha_1) \qquad\qquad (8)$$
$$\nu_1 = 1 - \tau^2/\kappa(\omega)^2,$$

where $k1n_,(\omega)$, is the fundamental wave vector in the sample.

As the result of solution of the system (2) one finds for α_1:

$$\alpha_{1\pm} = \pm(\nu_1^2 - \delta_1^2)^{½}/2. \qquad (9)$$

Introducing a similar parametrization for the second harmonic (α_2 and ν_2) one finds the next connection between the parameters of the fundamental wave and second harmonic:

$$\nu_2 = 1-(1-\nu_1)(1-\eta)/4, \qquad (10)$$

where $\eta=1-\varepsilon(\omega)/\varepsilon(2\omega)$ is the parameter determined by the frequency dispersion of dielectric susceptibility.

The solutions of the equations (4) for SH is a superposition of a particular solution of the inhomogeneous equations and the eigen solutions of the homogeneous equations with the wave vectors determined by the parameter ν_2. The wave vector in the particular solution is determined by zero value of the δ-functions arguments entering the right hand side of this equation. So for the wave vector in the particular solution one has:

$$k_{in} = -2\tau - \tau(\alpha_{1\pm} + \alpha_{1\pm}), \quad (11)$$

where in the right hand side of (11) all combinations of signs for $\alpha_{1\pm}$.are possible. The phase-matching condition of SHG demands coincidence of the particular solution wave vector with the wave vector of the eigen solution and may be presented in the following general form:

$$(1 - (k_{in}/\kappa)^2)\,(1 - (k_{in}+2\tau)^2/\kappa^2) - \delta_2^2 = 0 \quad (12)$$

where parameter δ_2 is determined by the second harmonic in the Fourier expansion of the Sm-C* dielectric tensor at the doubled fundamental wave frequency [11,17].The specific forms of the PM condition (12) are different for different nonlinear polarizations entering Eq.(4). The analysis shows that only n=0,2 in Eq.(5) result in phase-matched SHG for the fundamental wave being close to the RB edge. Therefore the consideration below will be restricted only by these values of n.

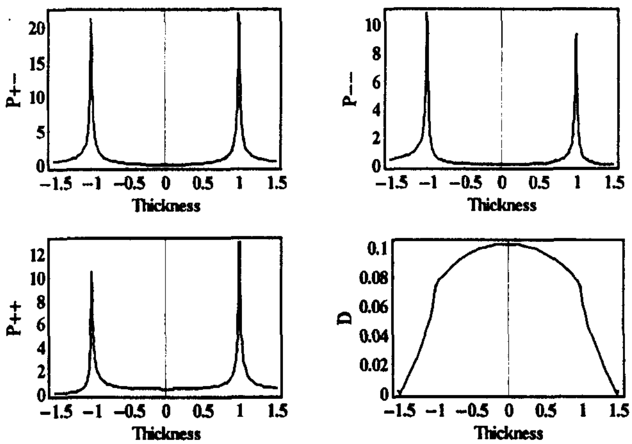

FIGURE 2 Nonlinear polarization and modulus of Eq. (12) (arbitrary units) versus ν_1/δ_1 (frequency or pitch variations) for the sample thickness l=20.

For n=2 the corresponding P_{+-} nonlinear polarization is due to the $\chi_{2\tau}$

term of the nonlinear susceptibility, gives $k_{in} = -2\tau$ and according to (12) results in the following PM condition for the fundamental wave:

$$\nu_1(1-\eta)+\eta=\delta_2^2, \qquad (13)$$

Taking into account typical values of η and δ_1 one concludes that this PM condition corresponds to the fundamental wave frequency being almost in the center of RB.

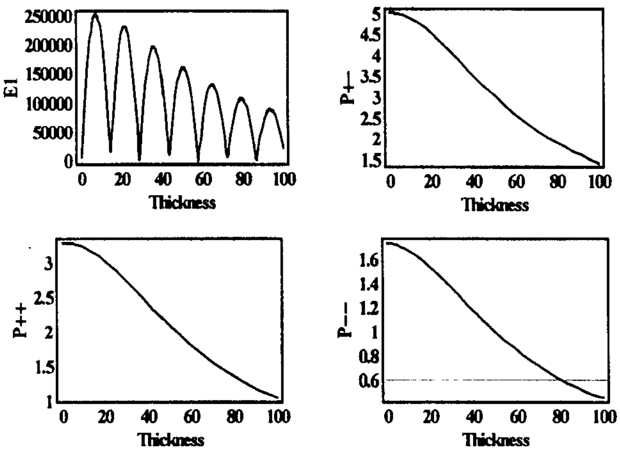

FIGURE 3 Nonlinear polarization and modulus of E_1 (arbitrary units) versus the sample thickness ($\nu_1/\delta_1 = 1.05$)

For n=0 the corresponding P_{++}, P_{--}, nonlinear polarizations are due to the χ_0 term of the nonlinear susceptibility, gives $k_{in} = -2\tau(1+\alpha_{1\pm})$ and according to (12) results in the following PM condition for the fundamental wave:

$$\nu_1(1-\eta)+\eta-(1-\nu_1)(1-\eta)((\alpha_{1\pm})^2+2\alpha_{1\pm})(1-(1-\nu_1)(1-\eta)(\alpha_{1\pm})^2)=\delta_2^2, \qquad (14)$$

This PM condition leads to $\nu_1 \approx \pm\delta_1$, what means that PM occurs close to the SR edge for the fundamental wave and an enhancement of SHG can be easily achieved. Eqs. (12-14) determine the value of ν_1 corresponding to the phase matched SHG. It may be the deviation of the pitch if the frequency ω is fixed or the deviation of the frequency from its Bragg value if the pitch is fixed. Because the parameters δ_1, δ_2, η in the real situation are small the phase matching condition (12) holds generally speaking not so far from the RB edge for the fundamental wave, i.e. in the ν_1 range where the nonlinear polarizations enhancement can reveal itself. The maximal SHG enhancement occurs if PM is reached exactly at the RB edge of the fundamental wave, i.e. for $\nu_1=\pm\delta_1$. However PM at this point can occur if only the Eq.(12) is satisfied at $\nu_1=\pm\delta_1$. It means that for a full scale enhancement the following relationship has to be satisfied:

$$\pm\delta_1(1-\eta)+\eta=\delta_2^2. \tag{15}$$

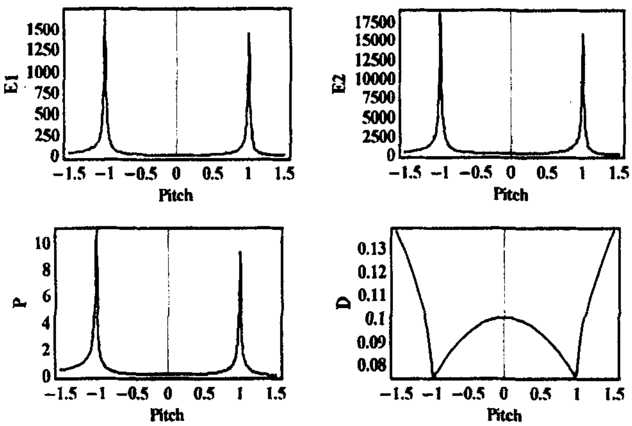

FIGURE 4 SH amplitude E_1, E_2, corresponding nonlinear polarization P_{--} and modulus of Eq. (12) (arbitrary units) versus ν_1/δ_1 for the sample thickness l=20.

SHG INTENSITIES

Solving Eqs (2,4) one finds for the SH amplitudes emanating from the input and output surfaces of the sample the following expressions:

$$E_1(z=L) = \{e_0 \exp(i(\alpha_{1\pm}+\alpha_{1\pm})l/2)+[e_0(\xi_+-\xi_-)+2ie_1\sin(\alpha_2 l)]/(\xi_-\exp(-i\alpha_2 l) -\xi_+\exp(i\alpha_2 l))\}/D$$

$$E_2(z=0) = \{e_1 + [e_1(\xi_+ - \xi_-) + 2ie_0\sin(\alpha_2 l)]/(\xi_-\exp(-i\alpha_2 l) -\xi_+\exp(i\alpha_2 l))\}/D \qquad (16)$$

where $\alpha_2=(\nu_2^2-\delta_2^2)^{1/2}/2$, and ν_2 is related to ν_1 by Eq.(10)

$$\xi_\pm = -\delta_2/[\nu_2 \pm\alpha_2], \quad D=(1-(k_{in}/\kappa)^2)(1-(k_{in}+2\tau)^2/\kappa^2)-\delta_2^2,$$

$$e_0 =-[(1-(k_{in}+2\tau)^2/\kappa^2)P_{-\tau} - P_\tau\delta_2], \; e_1=-[(1-(k_{in}/\kappa)^2)P_\tau - P_{-\tau}\delta_2], \qquad (17)$$

and $P_{-\tau}$ and P_τ, are determined by Eqs. (1,4,7).

Eq.(17) describes the dependence of the second harmonic amplitudes on ν_1 and the sample thickness.

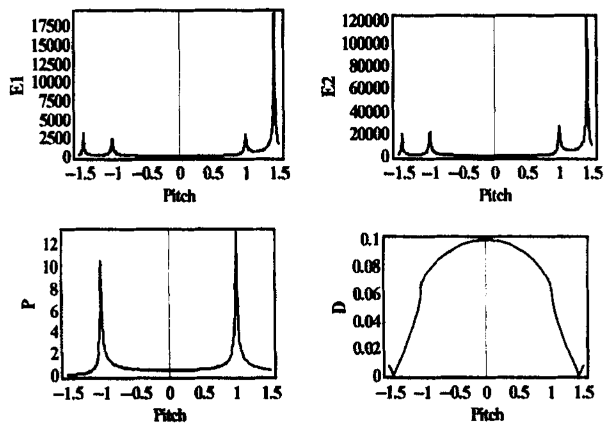

FIGURE 5 SH amplitude E_1, E_2, corresponding nonlinear polarization P_{++} and modulus of Eq. (12) (arbitrary units) versus ν_1/δ_1 for the sample thickness l=20.

CALCULATION RESULTS

To obtain a quantitative picture of the SGH enhancement for different cases of PM calculations were performed for the specific values of the parameters entering in the problem. The following values of the parameters were used in the calculations: $\delta_1=\delta_2=0.07$, $\eta=0.01$. It was also assumed that the dielectric constant out of the sample coincides with averaged dielectric constant for Sm-C*. The last assumption helps to rid off the SHG intensity beats connected with reflections at the dielectric boundaries.

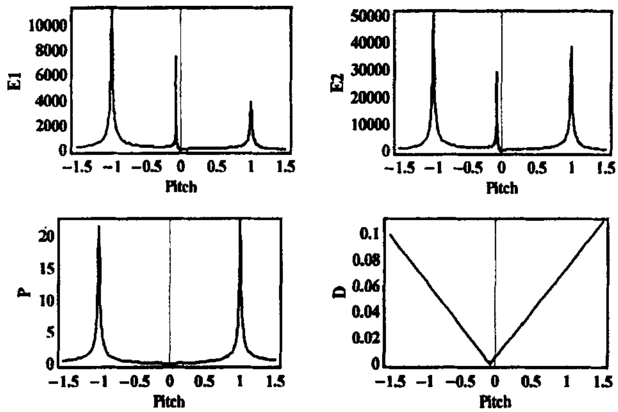

FIGURE 6 SH amplitude E_1, E_2, corresponding nonlinear polarization P_{+-} and modulus of Eq. (12) (arbitrary units) versus ν_1/δ_1 for the sample thickness l=20.

Figures 2,3, show behaviour of the nonlinear polarizations as functions of the deviation of the pumping wave from the Bragg condition, i.e. the dependence on parameter $\nu 1$ and the dependencies on the sample thickness, respectively. All nonlinear polarizations P++, P+- and P-- increase sharply at the RB edge of pumping wave. The sharpness of the maximum increases with increasing of the sample thickness. Maximal value of nonlinear polarization (see Fig.3) is reached for the finite sample thickness. Further increase of the sample thickness leads to decrease of the nonlinear polarization. Calculations of the SH amplitude for the different PM conditions(Figs.4-7) show that its maximum is located close to the RB edge for the fundamental frequency independently on the value of frequency dispersion and the corresponding SHG enhancement is due to the maxima of nonlinear polarizations at the SR edge with its frequency location not coinciding with the PM condition (The PM maxima corresponding to the P++ , P--,nonlinear polarizations are rather close to the SR edge). Usually the PM maxima of the SHG are less prominent than the corresponding maxima at the RB edge. However, if special choice of the parameters is satisfied and the PM condition coincides with the SR edge the enhancement of SHG is manifold magnified.

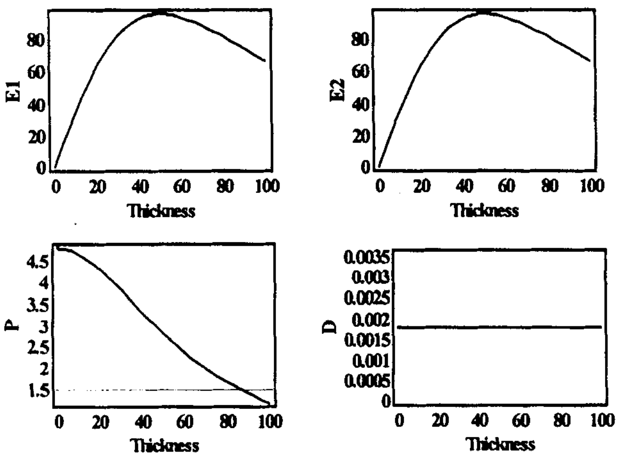

FIGURE 7 SH amplitude E_1, E_2, corresponding nonlinear polarization P_{+-} and modulus of Eq. (12) (arbitrary units) versus the sample thickness for ν_1/δ_1 =0.95.

CONCLUSION

The results obtained above demonstrate possibility of the enhanced nonlinear frequency conversion in Sm-C* if the fundamental wave is subjected to diffraction. More favourable for the enhancement is the homogeneous in space component of nonlinear susceptibility resulting in the PM conditions almost at the RB edge. The second space harmonic of the nonlinear susceptibility results in the PM conditions almost at the RB center, so the enhancement is also quite prominent however the maximal enhancement demands special relation between the Sm-C* parameters. The first space harmonic of the nonlinear susceptibility results in the PM conditions far away of the RB, so the enhancement of the SHG is practically absent for it.

Note that the SHG proceeds with unusual dependence on the parameters relevant to the problem, for example on the sample thickness. Result presented here relates to a simple case of polarization separation for light propagating along the helix however without any doubts qualitatively the same effects are present in the general situation when the polarization separation is not the case and the polarization properties of SHG are more sophisticated.

In the discussion above we assume that the PM is reached by changing of the pitch or frequency of the fundamental wave. However more practical, as shows the experiment with LC, is the first possibility to reach the effect without any changing of the fundamental wave frequency by variation of the LC temperature what in its turn leads to changing of the Sm-C* pitch.

Note also that the enhancement of nonlinear frequency transformation is not specific only of Sm-C* but is a very general phenomenon which exists in nonlinear periodical media of any nature [9, 10]. The LC, may be, are the best materials for studying this effect because there are lot of possibilities to change their periodicity and other parameters.

ACKNOWLEGEMENT

This work is supported by the INTAS grant N 30234.

References

[1] V.A. Belyakov and N.V. Shipov, *Phys. Lett.* **86A**, 94 (1981).
[2] V.A. Belyakov and N.V. Shipov, *Sov. Phys. JETP* **55**, 674 (1982).
[3] V.A. Belyakov and N.V. Shipov, *Sov. Tech. Phys. Lett.* **9**, 9 (1983).
[4] S.V. Shiyanovskii, *Ukr. Fiz. Zhurn*, **27**, 361 (1982).
[5] K. Kajikawa., T. Isozaki, H. Takezoe et al., *Jpn. J. Appl. Phys.* **31**. L679 (1992).
[6] T. Furukava, T. Yamada, K. Ishikawa et al., *Appl. Phys.* **B 60**, 485 (1995).
[7] J. Yoo, S. Choi, H. Hoshi et al., *Jpn. J. Appl. Phys.* **36**, L1168 (1997).
[8] V.A. Belyakov, *JETP Lett.*, **70**, 811 (1999).
[9] M. Scalora, M.J. Bloemer, A.S. Manka et al., *Phys. Rev.* A, **56**, 3166 (1997).
[10] J.W. Haus, R. Viswanathan, M. Scalora, et al., *Phys. Rev.* A, **57**, 2120 (1998).
[11] V.A. Belyakov, *Diffraction Optics of Complex-Structured Periodic Media* (Springer, NY, 1992).
[12] M. Copic and I. Drevensek-Olenik, *Liq. Cryst.* **21**, 233 (1996).
[13] I. Drevensek-Olenik and M. Copic, *Phys. Rev.* E, **56**, 581 (1997).
[14] S.V. Shiyanovskii, *SPIE Proc.*, **2795**, 2 (1996).
[15] V.A. Belyakov and M. Copic, *MCLC*, **328**, 151 (1999).

[16] D. Chang, H. Hoshi, K. Ishikawa et al., *MCLC*, **328**, 283 (1999).
[17] V.A. Belyakov and V.E. Dmitrienko, *Optics of Chiral Liquid Crystals*(Harwood Academic Publishers, Soviet Scientific Reviews, section A, V. 13, part 1 (1989)).