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

Theory of Second Harmonic Generation in Periodic Ferroelectric Materials

D. S. Moroi [a] & D. E. Zelmon [b]

[a] Liquid Crystal Institute, Kent State University, Kent, OH, 44242

[b] Wright Laboratory, Wright-Patterson AFB, OH, 45433

Version of record first published: 04 Oct 2006

Mol. Cryst. Liq. Cryst., 1997, Vol. 299, pp. 97–103
Reprints available directly from the publisher
Photocopying permitted by license only

THEORY OF SECOND HARMONIC GENERATION IN PERIODIC FERROELECTRIC MATERIALS

DAVID S. MOROI
Liquid Crystal Institute, Kent State University, Kent, OH 44242
DAVID E. ZELMON
Wright Laboratory, Wright-Patterson AFB, OH 45433

Abstract: Second harmonic generation is investigated in SmC^* liquid crystals as an example of periodic ferroelectric materials. The electric field is expanded in a Fourier series having the same periodicity as the ferroelectric material. If the nonlinear dielectric susceptibility is ignored, the Fourier components, or the amplitudes of the electric field in various modes, satisfy a five term vector recursion relation and the solutions are obtained by iteration for small tilt angles. The exact linear solutions are obtained for normal incidence and become identical with those for a cholesteric liquid crystal if the tilt angle becomes 90°. In the lowest order approximation, it is shown that the second harmonic field is given by a product of the linear solution multiplied by the leading term of the Fourier series which is calculated using the slowly varying envelope approximation.

INTRODUCTION

Recently, there has been considerable interest in optical materials with a large second order nonlinear susceptibility, $\chi^{(2)}$, for use in second harmonic generation (SHG) devices. Second harmonics can be generated effectively by the interaction of intense radiation with nonlinear optical materials such as potassium dihydrogen phosphate (KDP), lithium iodate (LiO_3), etc.[1], if the phase matching (PM) condition are satisfied.[2] It is also possible to obtain fairly efficient conversion in optical SHG by using quasi-phase matching (QPM) for nonlinear materials with a periodic structure.[3] QPM involves repeated reversal of the relative phase difference between the forced and free waves after an odd number of coherent lengths.[3] Because a nonlinear material with a periodic structure has one more parameter - the periodicity of dielectric function $\varepsilon(z)$ and nonlinear dielectric susceptibility $\chi^{(2)}(z)$ - than nonlinear crystals, there is a better chance of PM being achieved. Motivated by the latter idea, we present a theory of fairly efficient conversion in optical SHG for bulk continuous materials with spatially continuous periodic structures, in particular chiral smectic C liquid crystal (SmC*).[4,5]

LINEAR SOLUTIONS

We start with the wave equation in a nonlinear medium with a periodic structure

$$\nabla \times (\nabla \times \vec{E}) + \frac{\partial^2}{c^2 \partial t^2}(\varepsilon \cdot \vec{E}) = -\frac{\partial^2}{c^2 \partial t^2}\vec{P}_{NL} \tag{1}$$

Here c is speed of light in free space, ε the linear dielectric tensor of rank 2, and $\vec{P}_{NL}$ the nonlinear polarization of the periodic medium, which is given in detail in next section.

In what follows, a SmC^* has been chosen among various nonlinear periodic materials in our model calculation. Here we use a coordinate system in which the x-y plane is taken to be the plane boundary between a SmC^* and a substrate (isotropic homogeneous glass plate) or air and the z axis (normal to the smectic layers) to be positive going into the material.

The dielectric tensor of a SmC^* may be expressed as[4]

$$\varepsilon(z) = \sum_{m=-2}^{+2} \varepsilon(m) e^{imqz} \tag{2}$$

with $q = 2\pi/\Lambda$ and Λ being the pitch or periodicity of the material.

$$\varepsilon(0) = \begin{pmatrix} \varepsilon_{11} & 0 & 0 \\ 0 & \varepsilon_{11} & 0 \\ 0 & 0 & \varepsilon_{33} \end{pmatrix}, \tag{2a}$$

$$\begin{aligned} \varepsilon_{11} &= \frac{1}{2}[\varepsilon_1^{(P)} \cos^2\theta_t + \varepsilon_2^{(P)} + \varepsilon_3^{(P)} \sin^2\theta_t] \\ \varepsilon_{33} &= \varepsilon_1^{(P)} \sin^2\theta_t + (\varepsilon_1^{(P)} - \varepsilon_2^{(P)}) \cot^2\theta_t + \varepsilon_3^{(P)} \cos^2\theta_t , \end{aligned}$$

$$\varepsilon(\pm 1) = h_1 \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & \mp i \\ 1 & \mp i & 0 \end{pmatrix}, \quad h_1 = \frac{1}{4}(\varepsilon_3^{(P)} - \varepsilon_1^{(P)}) \sin 2\theta_t , \tag{2b}$$

$$\varepsilon(\pm 2) = h_2 \begin{pmatrix} 1 & \mp i & 0 \\ \mp i & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{2c}$$

$$h_2 = \frac{1}{4}(\varepsilon_1^{(P)} \cos^2\theta_t - \varepsilon_2^{(P)} + \varepsilon_3^{(P)} \sin^2\theta_t) \ ,$$

where θ_t is the tilt angle of the director of SmC^* to z-axis, $\varepsilon_1^{(P)}$, $\varepsilon_2^{(P)}$, and $\varepsilon_3^{(P)}$ are the principal values of $\varepsilon(z)$.

A linear solution to Eq. (1) with $\vec{P}_{NL} = 0$ is written in a Fourier series[5]

$$\vec{E}_L(\vec{r}, t) = \sum_{l=-\infty}^{+\infty} \vec{A}(l) e^{i(\vec{k}(l)\cdot\vec{r} - \omega t)} \tag{3}$$

where $\vec{A}(l) = \vec{A}(\vec{k}(l))$ is the complex amplitude of the lth Fourier component of the electric field and $\vec{k}(l)$ is the corresponding wavevector which is given by

$$\vec{k}(l) = \vec{k} + lq\hat{z} \tag{3a}$$

If x-z plane is chosen as the plane of incidence, we may write

$$\vec{k} = (k\sin\theta,\, 0,\, k\cos\theta) \ , \ \cos\theta = \hat{k}\cdot\hat{z} \tag{3b}$$

Substituting Eq. (3) into Eq. (1) with $\vec{P}_{NL} = 0$ and using the orthogonality properties of sinusoidal functions, we obtain the recursion relation for $\vec{A}(l)$

$$\Omega(l)\cdot\vec{A}(l) - (\frac{\omega}{c})^2 \sum_{\substack{m=-2 \\ m\neq 0}}^{+2} \varepsilon(m)\cdot\vec{A}(l-m) = 0 \tag{4}$$

where

$$\Omega(l) = (\vec{k}(l)\cdot\vec{k}(l))I - \vec{k}(l)\vec{k}(l) - (\frac{\omega}{c})^2\varepsilon(0) \tag{4a}$$

Although we do not present the derivation, a five term recursion relation may be found to determine the amplitude coefficients $A(l)$. Full account of these latter recursion relations and their solutions will be presented elsewhere.

It is, in general, difficult to obtain the solutions to Eq. (4) in closed analytical form. However, for small tilt angles (less than 12°), it is possible to obtain the solutions by iteration, provided that $\det\Omega(l)(l \neq 0)$ is not too small. (See Eqs. (2a)-(2b)). The solutions are

$$\vec{A}(l) = M(l)\cdot\vec{A}(0) \tag{5}$$

where for $l > 0$,

$$M(l+1) = G(l+1)[\varepsilon(1)M(l) + \varepsilon(2)M(l-1)] \tag{5a}$$

$$G(l) = \Omega(l)^{-1},\ M(0) = I$$

for $l < 0$,

$$M(l-1) = G(l-1)[\varepsilon(-1)M(l) + \varepsilon(-2)M(l+1)] \tag{5b}$$

and $\overrightarrow{A}(0)$ is the leading term of Fourier series representing the electric field. The wavevector $\overrightarrow{k}$ in the second order approximation is one of the eigenvalues which satisfy the secular equation

$$\det[\Omega(0) - \sum_{\substack{m=-2 \\ m\neq 0}}^{2} \varepsilon(m)M(-m)] = 0 \tag{6}$$

For the special case of normal incidence, we have obtained the exact solutions in closed analytical form for any tilt angle. We present the results without derivation

$$\overrightarrow{E}_{\pm}(\overrightarrow{r},t) = A_{\pm}\hat{x}_{\mp}\exp[i(\overrightarrow{k}_{\pm}\cdot\overrightarrow{r} - \omega t)] \tag{7}$$

where

$$A_{\pm} = \frac{1}{\sqrt{2}}(A_x \pm iA_y),\ \hat{x}_{\pm} = \frac{1}{\sqrt{2}}(\hat{x} \pm i\hat{y}) \tag{7a}$$

$$\overrightarrow{k}_{\bullet} = \overrightarrow{k} \pm \overrightarrow{q} = \frac{\omega}{c}\overrightarrow{\kappa}_{\pm} = \frac{\omega}{c}(\kappa \pm \delta)\hat{z},\ \delta = q/(\frac{\omega}{c}) \tag{8}$$

$$\kappa^2 = \varepsilon_{11} - \frac{2h_1^2}{\varepsilon_{33}} + \delta^2 \pm 2[(\varepsilon_{11} - \frac{2h_1^2}{\varepsilon_{33}})^2\delta^2 + (h_2 - \frac{h_1^2}{\varepsilon_{33}})^2]^{1/2} \tag{9}$$

It is readily proved that, for the special case of $\theta_t = \pi/2$ or $h_1 = 0$ and $\varepsilon_1^{(P)} = \varepsilon_2^{(P)}$, Eq. (9) reduces to a formula for the effective index of refraction for normal cholesteric liquid crystals.[6]

SECOND HARMONIC ELECTRIC FIELD

We study here SHG in SmC^*. Taking into account the Kleinman symmetry and C_2 point symmetry properties of the nonlinear dielectric susceptibility of a SmC^*, the $\chi^{(2)}$ may be written in dyadic form[7]

$$\chi^{(2)}(z) = \sum_{n=-3}^{3} \chi^{(2)}(n)e^{inqz} \tag{10}$$

Here

$$\chi^{(2)}(0) = 0 \tag{11}$$

$$\chi^{(2)}(1) = S[\chi^{(2)}_{--+}(1)\hat{x}_-\hat{x}_-\hat{x}_+ + \chi^{(2)}_{-zz}(1)\hat{x}_-\hat{z}\hat{z}] = \chi^{(2)}(-1)^* \tag{12}$$

$$\chi^{(2)}_{--+}(1) = \frac{i}{2\sqrt{2}}[\chi^{(2)}_{112}\cos^2\theta_t + 3\chi^{(2)}_{222} + \chi^{(2)}_{223}\sin^2\theta_t + \frac{1}{2}\chi^{(2)}_{123}\sin 2\theta_t] \tag{12a}$$

$$\chi^{(2)}_{-zz}(1) = \frac{i}{\sqrt{2}}[\chi^{(2)}_{112}\sin^2\theta_t + \chi^{(2)}_{233}\cos^2\theta_t - \frac{1}{2}\chi^{(2)}_{123}\sin 2\theta_t] \tag{12b}$$

where S implies a symmetric summation symbol defined by

$$S\hat{a}\hat{b}\hat{c} = \frac{1}{6}[\hat{a}(\hat{b}\hat{c} + \hat{c}\hat{b}) + \hat{b}(\hat{c}\hat{a} + \hat{a}\hat{c}) + \hat{c}(\hat{a}\hat{b} + \hat{b}\hat{a})]$$

We did not present $\chi^{(2)}(\pm 2)$ and $\chi^{(2)}(\pm 3)$, because they are very small compared with $\chi^{(2)}(\pm 1)$.

The electric fields for the fundamental ($\nu = 1$) and second harmonic ($\nu = 2$) in the nonlinear medium are described by

$$\vec{E}^{(\nu)}(\vec{r}, t) = \sum_{l=-\infty}^{\infty} \vec{E}^{(\nu)}(l; z) \exp[i(\vec{k}^{(\nu)}(l) \cdot \vec{r} - \omega^{(\nu)} t)] \tag{13}$$

where $\vec{E}^{(\nu)}(l; z)$ is the complex amplitude of the l th component of the electric field for $\nu = 1$ or 2 and $\vec{k}^{(\nu)}(l)$ is given by Eq. (3a) with substitutions $\vec{k} \to \vec{k}^{(\nu)}$ and $\omega \to \omega^{(\nu)}$. One may obtain extremely complicated coupled differential equations for $\vec{E}^{(1)}(z)$ and $\vec{E}^{(2)}(z)$ after laborious manipulation upon substitution of Eq. (13) into Eq. (1), and, in principle, they are solvable with the help of the method of multiple scales (MMS).[8] Using the slowly varying envelope approximation (SVEA), however, it can be shown that, in the lowest order approximation, the solution is

$$\vec{E}^{(\nu)}(l; z) = \vec{A}^{(\nu)}(l) E^{(\nu)}(z) / A^{(\nu)}(0) \tag{14}$$

where $\vec{A}^{(\nu)}(l)$ are the solutions of Eq. (4) with $\vec{k}$ and ω being replaced by $\vec{k}^{(\nu)}$ and $\omega^{(\nu)}$ and $E^{(\nu)}(z)$ is the complex amplitude of the zeroth Fourier component for the ν th harmonic which is calculated with SVEA. At this point, $E^{(\nu)}(z)$ is unknown. Substituting Eq. (14) into Eq. (13) first, substituting the resulting equation into Eq. (1), using SVEA, using the fact that $\vec{A}^{(\nu)}(l)$ is the linear amplitude and dropping $\chi^{(2)}(\pm 2)$ and $\chi^{(2)}(\pm 3)$ in $\vec{P}^{(\nu)}(z)$, we obtain

$$\frac{d}{dz} E^{(\nu)}(z) = i P^{(\nu)}(z) \tag{15}$$

where

$$P^{(\nu)}(z) = \left\{ \begin{array}{l} \eta_{\pm}^{(1)} e^{i\Delta_{\pm} z} E^{(1)}(z)^* E^{(2)}(z) \\ \eta_{\pm}^{(2)} e^{-i\Delta_{\pm} z} (E^{(1)}(z))^2 \end{array} \right. \tag{16}$$

with

$$\eta_{\pm}^{(1)} = \frac{(\omega/c)^2}{K^{(1)}} \vec{A}^{(1)*}(0) \cdot \chi^{(2)}(\mp 1, \omega) : \vec{A}^{(1)*}(0) \vec{A}^{(2)}(0) \tag{16a}$$

$$\eta_{\pm}^{(2)} = \frac{2(\omega/c)^2}{K^{(2)}} \vec{A}^{(2)*}(0) \cdot \chi^{(2)}(\pm 1, 2\omega) : \vec{A}^{(1)}(0) \vec{A}^{(1)}(0) \tag{16b}$$

$$K^{(\nu)} = 2Re(\vec{A}^{(\nu)*}(0) \times \vec{k}^{(\nu)}) \cdot (\vec{A}^{(\nu)}(0) \times \hat{z}) \tag{16c}$$

$$\Delta_{\pm} \equiv k_z^{(2)} - 2k_z^{(1)} \mp q \tag{16d}$$

The solutions to Eq. (15) are well-known and Jacobian elliptic functions. For the special case of perfect phase matching, however, they are given by

$$E^{(1)}(z) = A^{(1)}(0)\text{sech}\beta_\pm(z+z_0) \quad (17)$$

$$E^{(2)}(z) = i\frac{q}{\beta_\pm}\eta_\pm^{(2)}(A^{(1)}(0))^2\tanh\beta_\pm(z+z_0) \quad (18)$$

with

$$\beta_\pm \equiv [\eta_\pm^{(1)}\eta_\pm^{(2)}]^{1/2} \quad (17a)$$

Substituting Eq. (14) into Eq. (13), the formal solution in this approximation are given by

$$\vec{E}^{(\nu)}(\vec{r},t) = (A^{(\nu)}(0))^{-1}\vec{E}_L^{(\nu)}(\vec{r},t)E^{(\nu)}(z) \quad (19)$$

For normal incidence, $\vec{E}_L^{(\nu)}(\vec{r},t)$ in Eq. (19) should be replaced by the exact linear solution given by Eq. (7) with substitutions $A_\pm \to A_\pm^{(\nu)}$, $\vec{k}_\pm \to \vec{k}_\pm^{(\nu)}$, and $\omega \to \omega^{(\nu)}$.

CONCLUSION AND DISCUSSIONS

We have calculated the solutions for a SH field in SmC^*. The Type I PM conditions in this problem are given by

$$\vec{k}^{(2)} - 2\vec{k}^{(1)} \mp \vec{q} = 0\ , \quad (20)$$

as shown in Fig. 1, or they are written in terms of effective indices of refraction

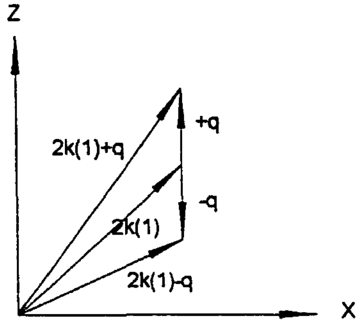

Fig. 1. Phase matching conditions for SHG in the chiral smectic C liquid crystal.

$$n^{(2)} \sin\theta^{(2)} = n^{(1)} \sin\theta^{(1)} \quad (20a)$$

$$n^{(2)} \cos\theta^{(2)} = n^{(1)} \cos\theta^{(1)} \pm \delta/2 \quad (20b)$$

where $\theta^{(\nu)} = \cos^{-1}(\widehat{k}^{(\nu)} \cdot \widehat{z})$, $\nu = 1, 2$.

It is readily seen from Eq. (20a) and (20b) that there is an additional good chance of fulfilling PM conditions if the materials have appropriate value of $\delta = q/(\frac{\omega}{c})$. Thus SH can efficiently be generated in such materials with reasonable value of $\chi^{(2)}(\pm 1)$. The formalism presented here is applicable to the nonlinear materials with the properties similar to SmC^{*}.

ACKNOWLEDGEMENT

Work was supported by AFOSR, The U.S. Air Force, under the 1995 University Resident Research Program.

REFERENCES

1.R. C. Eckart, et al, IEEE J. Q. E., QE26, 922-33 (1990).

2.N. Bloembergen, Nonlinear Optics, pp. 88-90. Benjamin, New York 1965.

3.M. M. Fejer, et al, IEEE J. Q. E. QE28, 2631-54 (1992).

4.S. A. Pikin, et al, in Ferroelectric Liquid Crystals, Vol. 7, pp. 268-71. G. W. Taylor, Ed., Gordon & Breach, Philadelphia, 1991.

5.V.A. Belyakov, Diffraction Optics of Complex Structure Periodic Media, Springer-Verlag, New York, 1992.

6.P. G. de Gennes & J. Prost, The Physics of Liquid Crystal, p. 273. Oxford, 1993.

7.R. G. Petschek, Case Western Reserve Univ., private communication.

8.N. Amarasinghe, D.S. Moroi and D. Zelmon, ALCOM Proc. Vol. 4, pp. 69-75, 1992. D. S. Moroi, Technical Report, Grant No. AFOSR-91-202, July 1993.