Contribution to the theory of nonlinear-optical frequency conversion in cholesteric liquid crystals

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Nonlinear-optical frequency conversion in a cholesteric liquid crystal is investigated theoretically under conditions of selective reflection of the generated radiation, with the third harmonic as the example. It is shown that when the harmonic is selectively reflected the efficiency of nonlinear frequency conversion can increase strongly and reach a maximum at the boundary of the selective-reflection region. The requirements that the parameters of the cholesteric and of the pump wave must be satisfied to ensure maximum nonlinear frequency conversion efficiency are indicated. Third-harmonic generation is described in the given pump-wave approximation for collinear geometry and it is shown that the corresponding growth of the increase of the harmonic is described by the relation $\delta\omega_e|\omega-\omega_e|^{-1}$ (ω and ω_e are the frequencies of the harmonic and of the selective-reflection boundary, and δ is the dielectric anisotropy of the cholesteric) and has a limiting value proportional to the fourth power of the sample thickness.

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INTRODUCTION

The study of nonlinear optical phenomena in liquid crystals (LC) is of considerable interest both for the physics of LC and for nonlinear optics itself and its applications.[1-4]

A thorough theoretical and experimental investigation of the nonlinear frequency conversion in cholesteric liquid crystals (CLC), using third-harmonic generation as an example, was carried out by Shen and Shelton.[1] Their experimental results were confirmed by subsequent investigations.[2] Principal attention in the cited papers was paid, however, to the study of more varied (compared with a homogeneous medium) possibilities of synchronous conversion of frequency, which are due to periodicity of the dielectric characteristics of the cholesterics. In addition, the analysis pertained to the case of a collinear geometry of third-harmonic generation, or else was limited to conditions under which neither the pumping wave nor the harmonic underwent strong diffractive scattering. The main content of the present paper is a study of nonlinear third-harmonic generation under conditions when it is diffracted by the periodic structure of the cholesteric, in which, just as in ordinary periodic media, a more substantial increase of the efficiency of the nonlinear frequency conversion can be obtained.[5]

In the given-pump-field approximation, we analyze the angular, frequency, and polarization characteristics, as well as those integrated over the frequency width and over the divergence angle of the pump wave, of the nonlinearly generated harmonics under conditions when they are diffracted by the periodic structure of the cholesteric. The conditions that must be satisfied by the parameters of the cholesteric and by the pump radiation to realize the maximum increase of the effectiveness of nonlinear conversion of the frequency in the cholesteric, compared with a homogeneous medium, are indicated. We obtain the dependence of the efficiency of frequency conversion on the deviation of the experimental conditions from the optimum, and indicate the deviation limits within which the increase of the frequency-conversion effectiveness is still substantial.

COLLINEAR GEOMETRY

General analysis. We consider nonlinear frequency conversion in a cholesteric for the case of propagation of the pump wave along the cholesteric axis, assuming for the sake of argument that we are dealing with third-harmonic generation in a planar cholesteric layer, i.e., in a sample whose surfaces are perpendicular to the cholesteric axis (the z axis) (see Fig. 1, putting $\theta=\pi/2$).

In the approximation when the pump field $\mathbf{E}(\omega)$ is given, linear generation of the third harmonic is described by the equation

$$\left[\frac{\partial^2}{\partial z^2}+\left(\frac{3\omega}{c}\right)^2\hat{\varepsilon}(z,3\omega)\right]\vec{\mathscr{E}}(z,3\omega)=-4\pi\left(\frac{3\omega}{c}\right)^2\vec{\mathscr{P}}^{(3)}(z,3\omega), \quad (1)$$

where $\hat{\varepsilon}$ is the dielectric tensor of the cholesteric, and $\vec{\mathscr{P}}^{(3)}$ is the nonlinear polarization vector expressed in terms of the cubic nonlinear susceptibility $\hat{\chi}^{(3)}(z,3\omega)$:

$$\vec{\mathscr{P}}^{(3)}(z,3\omega)=\hat{\chi}^{(3)}(z,3\omega)\mathbf{E}(z,\omega)\times\mathbf{E}(z,\omega)\mathbf{E}(z,\omega).$$

Here and elsewhere the quantities marked by the index (3) pertain to the frequency 3ω.

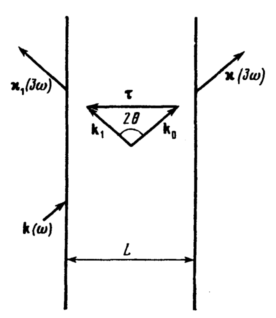

FIG. 1. Illustrating the geometry of frequency conversion under selective-scattering conditions.

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Just as in linear optics of cholesterics,[3] we seek the particular solution of the inhomogeneous equations (1) in the form

$$\vec{\mathscr{E}}(z,\ 3\omega)=e^{-3i\omega t}[E^+\hat{n}_+\exp(i\tilde{K}^+z)+E^-\hat{n}_-\exp(i\tilde{K}^-z)],\qquad (2)$$

where $\hat{n}_\pm=(\hat{x}\pm i\hat{y})/\sqrt{2}$ are the unit vectors of the circular polarizations, and the wave vectors $\tilde{K}^\pm$ satisfy the relation $\tilde{K}^+-\tilde{K}^-=\tau$, where $\tau=4\pi/p$ is the vector of the reciprocal lattice of the cholesteric and p is the pitch of the helix. As a result we obtain for the amplitudes $E^\pm$ the following system of equations

$$\begin{aligned}&[1-(\tilde{K}^+/\varkappa_3)^2]E^++\delta E^-=\sum_{sj_1j_2j_3}\mathscr{P}^{+lmns}_{j_1j_2j_3}\delta(\tilde{K}^+-K_{j_1}{}^l-K_{j_2}{}^m-K_{j_3}{}^n-s\tau),\\&\delta E^++[1-(\tilde{K}^-/\varkappa_3)^2]E^-=\sum_{sj_1j_2j_3}\mathscr{P}^{-lmns}_{j_1j_2j_3}\delta(\tilde{K}^--K_{j_1}{}^l-K_{j_2}{}^m-K_{j_3}{}^n-s\tau),\end{aligned}\qquad (3)$$

where $\varkappa_3=3\omega[\bar{\varepsilon}(3\omega)]^{1/2}/c$, $\bar{\varepsilon}_3=\bar{\varepsilon}(3\omega)$ is the average value of the dielectric constants of the cholesteric at the frequency 3ω; $l,\ m,n=\pm,\ s=0\pm1\pm2\pm3\ldots,K_j^\pm$ are the wave vectors in an expansion similar to (2) of the eigensolutions of the system (1) at the frequency ω ($j=1-4$ number the eigensolutions[3]), and $\mathscr{P}^{\pm lmns}_{j_1j_2j_3}$ are the Fourier components of the expansion in the nonlinear-polarization series

$$\mathscr{P}^{(3)}_{(z,3\omega)}=-\frac{\varepsilon_3}{4\pi}\sum\hat{n}_q\mathscr{P}^{qlmns}_{j_1j_2j_3}\exp[i(K_{j_1}{}^l+K_{j_2}{}^m+K_{j_3}{}^n+s\tau)z],\quad q=\pm.$$

The harmonic is most efficiently generated, as is known, under synchronism conditions, to satisfy which the quantities $\tilde{K}^\pm$ must satisfy the dispersion equation at the frequency 3ω and simultaneously cause the argument of at least one δ-function in the right-hand side of (3) to be zero. In the customarily employed approximation, where the local linear and nonlinear dielectric properties of the cholesteric are identical with those of the nematic,[1] i.e., for a uniaxial crystal, the synchronism conditions take the form

$$k^\pm(3\omega)=k^\pm(\omega)+k^\pm(\omega)+k^\pm(\omega),\qquad (4)$$

where arbitrary combinations of the signs are possible in (4), and in addition to k^m it is possible to have also $-k_m$ in (4), while the values of $k^m (m=\pm)$ for the frequencies ω and 3ω are defined by the relation

$$(k^\pm)^2=\varkappa^2+\tau^2/4\pm\varkappa[\tau^2+\delta^2\varkappa^2]^{1/2}.\qquad (5)$$

Under synchronism conditions the $\tilde{K}^\pm$ are expressed in the following manner in terms of $k^\pm(3\omega)=k_3^\pm$

$$\tilde{K}^+=k_3{}^\pm+\tau/2,\quad \tilde{K}^-=k_3{}^\pm-\tau/2.$$

We note that allowance for the difference between the local linear or nonlinear dielectric characteristics of the cholesteric and the characteristics of the nematic, i.e., allowance for the spatial dispersion[6] in the absence of an inversion center in the cholesteric, leads to a generalization of the synchronism conditions (4), wherein there is added in the right-hand side of (4) a term $s\tau$, where $s=\pm1,\ \pm2,\ldots$, as well as a term due to molecular gyrotropy.[7] However, in view of the usual neglect of the molecular gyrotropy and absence of central symmetry, i.e., of the smallness of the aforementioned difference, the most intense generation corresponds to the synchronism conditions (4), which will therefore be analyzed below.

Synchronism under diffraction conditions. Concentrating hereafter mainly on the influence of diffraction of the harmonic in the cholesteric on the efficiency of the nonlinear frequency conversion and using the fact that circularly polarized light (having the same sign as the cholesteric helix) is diffracted in propagation along the cholesteric axis, we shall investigate the singularities of the generation of a harmonic having the same diffraction of the circular polarization. Recognizing that the diffraction scattering is experienced by the eigensolution corresponding to k_3^-, we obtain the synchronism conditions for the generation of the harmonic with the diffracting polarization by substituting in the left-hand side of (4) only the quantity k_3^-.

For an arbitrary pump-wave frequency ω we can reach satisfaction of the synchronism conditions by selecting the corresponding pitch of the cholesteric helix, the synchronous value of which turns out to depend on the frequency dispersion of the dielectric constant $\bar{\varepsilon}$ and on the anisotropy δ. The synchronism conditions are reached in the general case outside the region of selective reflection of the harmonic, and third-harmonic generation for this situation was investigated by Shelton and Shen.[1]

Of special interest is a situation in which the synchronism is reached near the region of selective reflection. We assume that the third-harmonic frequency tends to one of the edges of the region of the selective reflection, i.e., $k_3^-=\varkappa_3\xi$, where $\xi\ll\delta$, and a relation corresponding to the exact coincidence of the frequency with the boundary of the region of selective reflection is almost satisfied, i.e.,

$$\tau=2\varkappa_3[1+\xi^2\pm\delta(1+4\xi^2/\delta^2)^{1/2}]^{1/2}.\qquad (6)$$

We find from (4) that the only synchronism condition that can be satisfied in this case is the equality

$$2k^-(\omega)+k^+(\omega)=k^-(3\omega)=\xi\varkappa_3.\qquad (7)$$

Using for $k^\pm$ and τ expressions (5) and (6), respectively, we find from (7) that the condition for obtaining synchronism at the boundary of the selective-reflection region of the harmonic ($\xi=0$) is the following relation between the frequency dispersion and the anisotropy of the dielectric properties of the cholesteric

$$d=\{41-40[1+(3\delta_1/8)^2]^{1/2}\}/(1-25\delta_1{}^2/9),\qquad (8)$$

where $d=\bar{\varepsilon}_1/(1\pm\delta_3)\bar{\varepsilon}_3$, the indices 1 and 3 mark values pertaining respectively to the frequencies ω and 3ω. Relation (3) reduces approximately to $\bar{\varepsilon}_3=\bar{\varepsilon}_1(1\pm\delta_3)$.

Thus, assuming the frequency dependences of the quantities in (8) to be known, we can obtain from this relation the frequency ω, and by the same token also the pitch of the cholesteric helix for which the frequency of the harmonic turns out to be exactly at the boundary of the region of selective reflection. From the practical point of view, however, this means that if, as usual, the frequency ω (the laser frequency) is fixed, it is necessary to choose specially the parameters of the cholesterics if the synchronism condition is to coincide with the boundary of the region of selective reflection.

Solution of the boundary-value problem. In the planar cholesteric layer considered by us, the field of the harmonic is represented as a superposition of the particular solution of the system (1), (3) with the eigenwaves of frequency 3ω. The coefficients in this superposition are obtained from the boundary conditions. By solving the formulated boundary-value problem we obtain the following expressions for the amplitudes of the harmonic fields that emerge through the entry and exit surfaces of the layer:

$$E^+=D^{-1}\left[D^+ +\frac{D^+(\xi_+ +\xi_-)\exp[-i(\bar{K}^+-\tau/2)L]+2iD^-\sin(k_3^-L)}{\xi_-\exp(-ik_3^-L)-\xi_+\exp(ik_3^-L)}\right],$$
$$E^-=D^{-1}\left[D^- +\frac{D^-(\xi_+ -\xi_-)\exp[i(\bar{K}^+-\tau/2)L]+2i\xi_+\xi_-D^+\sin(k_3^-L)}{\xi_-\exp(-ik_3^-L)-\xi_+\exp(ik_3^-L)}\right]; \quad (9)$$
$$D=[1-(\bar{K}^+/\varkappa_3)^2][1-(\bar{K}^-/\varkappa_3)^2]-\delta_3^2,$$
$$D^+=\mathscr{P}^+_{lmn}[1-(\bar{K}^-/\varkappa_3)^2]-\delta_3\mathscr{P}^-_{lmn},\quad D^-=\mathscr{P}^-_{lmn}[1-(\bar{K}^+/\varkappa_3)^2]-\delta_3\mathscr{P}^+_{lmn};$$
$$\xi_\pm=[\varkappa_3^{-2}(\tau/2\pm k_3^-)^2-1]/\delta_3.$$

The indices $l, m, n = \pm$ correspond to the indices that enter in the synchronism conditions (4) and (7), while the $\mathscr{P}^\pm_{lmn}$ are defined in the Appendix [see (A3)]. We note that expressions (9) were obtained neglecting reflections from the sample boundary, i.e., they pertain in fact to the case of small δ and a small difference between $\bar{\varepsilon}$ and the dielectric constant of the external medium.

As follows from (9), the amplitudes of the third harmonic as functions of the small difference between the frequency 3ω and the boundary ω_e of the selective-reflection region, defined by relation (6) at $\xi = 0$, oscillates strongly at or close to synchronism conditions. To verify this, we obtain, e.g., the expressions for the synchronous values of the amplitudes $E^\pm$ on the sample surface:

$$E^\pm=\pm i\frac{\varkappa_3^2L(\xi_+-\xi_-)\exp(ik_3^-L)D^\pm}{16k_3^-[\xi_-\exp(-ik_3^-L)-\xi_+\exp(ik_3^-L)]}. \quad (10)$$

It follows from (10) that when the synchronism is reached near the boundary of the selective reflection for the harmonic, i.e., at $|k_3^-/\varkappa_3| < \delta_3$, the efficiency of linear conversion of the frequency can greatly exceed the corresponding efficiency outside the diffraction conditions. Indeed, the expression (10) for the amplitudes $E^\pm$ undergoes strong oscillations (Fig. 2) as a function of the synchronous value of the quantity k_3^-, being proportional at the minima, as usual, to the sample thickness, and at the maxima to the square of the thickness.

Extremal conversion efficiency. As follows from (10), the maxima of the amplitudes $E^\pm$ are reached at values $k_3^- = 2k^-(\omega) + k^+(\omega) = \pi s/L$, ($s$ is an integer) and are equal to

$$E^\pm_{max}=\pm i(P^+-P^-)\frac{\delta(\varkappa_3L)^2}{8\pi s},\quad E^\pm_{min}\sim(P^+-P^-)\varkappa_3L, \quad (11)$$

where $P^\pm = P^\pm_{+--} + P^\pm_{-+-} + P^\pm_{--+}$, and the $P^\pm_{lmn}$ are given in the Appendix.

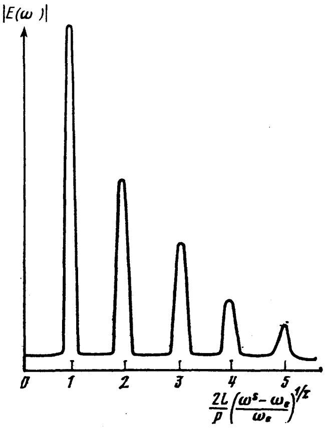

FIG. 2. Qualitative dependence of the harmonic amplitude on the synchronism frequency near the boundary of the selective reflection.

It is seen from (10) and (11) that the maximum value of at least one of the amplitudes, E^+ or E^-, is approximately equal to the amplitude outside the diffraction. The minimum value corresponds to an increase of the linear-conversion efficiency in the region of noticeable diffractive scattering of the harmonic, and outside this region $|k_3^-/\varkappa_3| > \delta$ it goes over into the known expressions which are proportional to $\varkappa_3 L$ (Ref. 1). It follows from (10) that the maxima of the generation efficiency are reached at the following values of the parameter $\nu = (3\omega - \omega_e)/\omega_e$ that describes the position of the frequency of the harmonic outside the boundaries $\omega_e = c\tau[\bar{\varepsilon}_3(1 \pm \delta_3)]^{-1/2}/2$ of the region of selective reflection:

$$\nu=\frac{1}{\delta}\left(\frac{2\pi s}{\tau L}\right)^2=\frac{1}{\delta}\left(\frac{ps}{2L}\right)^2. \quad (12)$$

It is assumed here that for the harmonic frequency 3ω satisfying the condition (12) the synchronism condition is also satisfied. From a comparison of (11) and (12) it is seen that the maximum generation intensity decreases, with increasing distance of the synchronous generation frequency from the boundary of the region of selective reflection, like

$$I^{max}_{(3\omega)}=I_0\frac{\delta\omega_e}{|3\omega-\omega_e|}, \quad (13)$$

where the frequency 3ω assumes discrete values determined by Eq. (12), and I_0 is a quantity approximately equal to the intensity of the synchronous generation of the harmonic in the same sample far from the selective-reflection region.

Relation (13) can be represented in a different form if it is assumed that the light frequency is fixed, while the variable is the pitch of the helix, and the synchronism near the boundary of the selective reflection is reached by varying the pitch:

$$I^{max}(\tau)=I_0\frac{\delta\tau_e}{|\tau-\tau_e|}, \quad (14)$$

where $p_e = 4\pi/\tau_e$ is the helix pitch corresponding to equality of the frequency 3ω to the boundary of the selective-reflection zone [see (6)], while the discrete values of τ are determined by Eq. (12) in which 3ω is replaced by τ and ω_e is replaced by τ_e.

We point out that the amplitude of the harmonic is proportional to $\delta_3(\varkappa_3 L)^2$ near the selective-reflection region not only when the determinant D of the system (3) is exactly equal to zero, and in particular the quantity when

$$\bar{K}^+ - \tau/2 = 2k^-(\omega) + k^+(\omega)$$

coincides with k_3^{-1}, but also near these conditions at

$$2k^-(\omega) + k^+(\omega) = \pi q/L, \quad k_3^- = \pi n/L.$$

In this case the amplitudes at the maxima reach

$$E^\pm = \frac{(P^+ - P^-)\delta(\varkappa_3 L)^2[1 - e^{i(n+q)\pi}]}{(2\pi)^2(n^2 - q^2)}, \tag{15}$$

where n is an integer and q is a number of opposite parity. It is seen from (11) and (14) that the absolute maximum of the intensity of the harmonic ($q = 0, n = \pm 1$) is reached practically on the boundary of the selective-reflection region. In the case of exact satisfaction of the synchronism conditions (7), the maximum corresponds to the value $s = 1$ in (11), meaning a very small deviation of the frequency from the boundary of the selective-reflection region.

Conditions for increase of efficiency. It is natural to seek the conditions under which the maximum harmonic amplitude is obtained in a real experiment if the dispersion differs from the limiting value (8). Bearing in mind the experimental possibility of easily varying the pitch of the cholesteric, we assume in this search that the free parameter of the problem is in fact the pitch of the helix, i.e., τ, while the remaining parameters, particularly the frequency ω, are fixed. By varying τ we can, e.g., satisfy the synchronism conditions (7), which now no longer correspond to the boundary ω_e of the region of selective excitation, or else make the frequency of the harmonic coincide with ω_e, or more accurately with the position of the first maximum, deviating thereby from the synchronism conditions. The first possibility may mean that the amplitude of the harmonic is described by Eq. (11) with $|s| \geq 1$, while the second means that it is described by Eq. (15) with $n = \pm 1$ and $q = 0$. An analysis of expressions (9)–(15) shows that if $\delta^{-1}(p/2L)^2$ is less than $\nu = (\tau^s - \tau_e)/\tau_e$, where τ^s corresponds to the synchronous pitch of the helix, and

$$\tau_e = (6\omega/c)[\bar{\varepsilon}_3(1 \pm \delta_3)]^{1/2},$$

an absolute maximum of the harmonic amplitude, proportional to $\delta_3(\varkappa_3 L)^2$, is reached under the synchronism conditions (7) and is determined by expression (11).

The maximum values of the intensity of the synchronous generation, determined by formulas (11), (13), and (15), pertain to a strictly monochromatic wave. An effect of the same order for a wave with finite frequency line width $\Delta\omega/\omega$ is reached if this line width is less than the frequency width of the maxima of the harmonic generation, which turns out to be of the order of $(\pi s/\delta)^2 \times (\varkappa_3 L)^{-3}$ and determines the "sharpness" of the synchronism under diffraction conditions.

On the other hand, if the line width is of the order of the frequency period of the oscillation of the amplitudes of the harmonic in (10), i.e., $\sim s/\delta(\varkappa_3 L)^2$, then the generation efficiency turns out to be less than the maximum, and is described by the squared modulus of (10) averaged over the frequency width of the line, and is determined by

$$I \sim I_0 \left(\frac{\delta\tau_e}{|\tau - \tau_e|}\right)^{1/2} = I_0 \left(\frac{\delta\omega_e}{|3\omega - \omega_e|}\right)^{1/2}, \tag{16}$$

where ω is the meaning of the frequency position of the line center and can vary continuously, satisfying the condition

$$|(3\omega - \omega_e)/\omega_e| \geq (p/2L)^2/\delta.$$

It is seen from (16) that the increase in the efficiency of the nonlinear conversion of the frequency takes place if the deviation of the frequency 3ω from ω_e and the pump line width do not exceed δ_3.

In analogy with the foregoing, we can write down the amplitudes of the harmonic field without assuming that the dielectric anisotropy δ is small and that $\bar{\varepsilon}_3$ is equal to the dielectric constant ε_e of the external medium. In this case an important role can be played by the reflection of the fields from the boundaries of the cholesteric layer and the ensuing transformation of one circular polarization into the other. The polarization of the harmonic that emerges from the cholesteric then turns out to be generally speaking elliptic. Without writing down the cumbersome general expressions for the field of the harmonic, we confine ourselves only to expressions for the intensities of the circularly polarized components of the harmonic field at the exit from the cholesteric at the maximum frequency conversion efficiency [$q = 0, n = \pm 1$ in (15)]:

$$|E_\pm^+|^2 = |E_\pm^-|^2 = (P^+ - P^-)^2\delta_3^2(L\varkappa_3^2/\pi k_3^+)^4[1 \pm (1 \pm \delta_3)^{1/2}]^2, \tag{17}$$

where the $P^\pm$ are defined in (11), and we put also $\bar{\varepsilon}_3 = \varepsilon_e$.

NONLINEAR FREQUENCY CONVERSION IN OBLIQUE INCIDENCE OF THE LIGHT

We proceed now to consider third-harmonic generation in cholesterics when the pump wave propagates at an angle to the cholesteric axis. As for the physical aspect of the problem, the harmonic-generation picture remains here qualitatively similar to the case of propagation of light along the cholesteric axis, but the polarization characteristics of the generated waves turn out to be in the general case to be more complicated, owing to the more complicated linear optics of the cholesterics for this case. An additional factor that may play an important role in experiment is that satisfaction of the synchronism conditions at a fixed frequency ω of the pump wave can be obtained generally speaking not only by changing the characteristics of the cholesteric, but also by changing the pump-wave propagation of direction.

Assuming that the pump-wave propagation direction is arbitrary, and that the field of the harmonic constitutes a superposition of two plane waves (Fig. 1)

$$\vec{\mathscr{E}}(\mathbf{r}, 3\omega) = e^{-3i\omega t}[\mathbf{E}_0 e^{i\mathbf{k}_0\mathbf{r}} + \mathbf{E}_1 e^{i\mathbf{k}_1\mathbf{r}}], \quad \mathbf{k}_1 = \mathbf{k}_0 + \boldsymbol{\tau}$$

we obtain from Maxwell's equations the following system:

$$[\hat{\varepsilon}_0 - (\mathbf{k}_0 c/3\omega)^2]\mathbf{E}_0 + \hat{\varepsilon}_{-1}\mathbf{E}_1 = \sum_{j_s} \vec{\mathscr{P}}^{0lmns}_{j_1j_2j_3}\delta(\mathbf{k}_{lj_1} + \mathbf{k}_{mj_2} + \mathbf{k}_{nj_3} + s\boldsymbol{\tau} - \mathbf{k}_0),$$
$$\hat{\varepsilon}_1\mathbf{E}_0 + [\hat{\varepsilon}_0 - (\mathbf{k}_1 c/3\omega)^2]\mathbf{E}_1 = \sum_{j_s} \vec{\mathscr{P}}^{1lmns}_{j_1j_2j_3}\delta(\mathbf{k}_{lj_1} + \mathbf{k}_{mj_2} + \mathbf{k}_{nj_3} + s\boldsymbol{\tau} - \mathbf{k}_1), \quad (18)$$

where $\hat{\varepsilon}_0$ and $\hat{\varepsilon}_{\pm 1}$ are the only nonzero Fourier components of the Taylor expansion of the dielectric tensor of the cholesteric, $\mathbf{k}_{0j}$ and $\mathbf{k}_{1j}$ are the wave vectors in the eigensolutions of Maxwell's equations at the pump-wave frequencies, $j = 1-4$, $\vec{\mathscr{P}}^{0,1lmns}_{j_1j_2j_3}$ are the Fourier components of the expansion of the nonlinear polarization of the form used in (3), $l, m, n = 0.1$, $s = 0 \pm 1 \pm 2 \pm 3 \ldots$. The explicit form of the matrix and of the determinant of the system (18) are given, e.g., in Ref. 3.

Just as for the collinear geometry, we find that the synchronism conditions are

$$\mathbf{k}_{0j}(3\omega) = \mathbf{k}_{lj_1}(\omega) + \mathbf{k}_{mj_2}(\omega) + \mathbf{k}_{nj_3}(\omega) + s\boldsymbol{\tau}. \quad (19)$$

The synchronism conditions (18) can be satisfied, depending on the concrete experimental conditions, both for cases when the field of the waves can be represented with sufficient accuracy by a single plane wave,[8] and when the diffraction of light is substantial and it is necessary to take into account in the eigensolutions their difference from plane waves.

Just as in collinear generation, the more interesting situation is the one in which the harmonic experiences diffraction by the periodic structure of the cholesteric. In this case, at all permissible values of s in the synchronism condition (19), the character of the harmonic generation differs from the case of a homogeneous medium. Generally speaking, for any s, contributions to the generation are made both by the spatially homogeneous and spatially inhomogeneous components of the nonlinear susceptibility. However, just as in the collinear case, the maximum efficiency is reached when the synchronism frequencies (directions) coincide with the edge of the selective-reflection band. In this case the generation intensity in the directions $\mathbf{k}_0(3\omega)$ and $\mathbf{k}_1(3\omega)$ turn out to be of the same order.

The sought solution of the system (18) constitutes a sum of the partial solution of the inhomogeneous system with superposition of the eigensolutions of the homogeneous problem, in which the coefficients are determined from the boundary conditions.

Without writing out here in explicit form the cumbersome solution in question, which, just as for the collinear geometry, can be obtained in standard fashion in analogy with the homogeneous problem,[3,9] we discuss first in greater detail the synchronous generation conditions.

Recognizing that $\mathbf{k}_{0j}(3\omega)$ in (19) satisfies the dispersion equation, we can write the synchronism condition in the cholesteric, i.e., the condition for the vanishing of the determinant of the system (18), in the form

$$D \equiv (\eta^2 - m^2)(\alpha^2 - m^2) - (\eta - m^2)(\alpha - m^2) = 0,$$
$$\eta = [1 - k_0^2/\varkappa_3^2(1 - \delta\cos^2\theta/2)](1 + m)/\delta, \quad (20)$$
$$\alpha = [1 - k_1^2/\varkappa_3^2(1 - \delta\cos^2\theta/2)](1 + m)/\delta, \quad m = \cos^2\theta/(1 + \sin^2\theta),$$

where k_0 and k_1 are defined in (18).

From the parameter α, which is not connected with the concrete geometry of the experiments, it is convenient to change over to the parameter Δ (see Ref. 3), which describes the optical properties of a planar cholesteric layer as a function of the boundary conditions. For a fixed frequency ω we have

$$\Delta = 2(\theta - \theta_B)\sin 2\theta/\delta(1 + \sin^2\theta), \quad \sin\theta_B = \tau/2\varkappa_3(1 - \delta\cos^2\theta/4),$$

and for a fixed angle θ (see Fig. 1)

$$\Delta = 4(3\omega - \omega_B)\sin^2\theta/\delta\omega_B(1 + \sin^2\theta), \quad \omega_B = c\tau/2\bar{\varepsilon}^{1/2}(1 - \delta\cos^2\theta/4).$$

The connection between the parameters α and Δ for the planar case are determined by the relation $\Delta = (\alpha + \eta)/2$, and by substituting in the latter the synchronous values of α, which are determined by the roots of Eq. (20), we obtain explicit expressions for the parameter Δ under the synchronism conditions:

$$\Delta = \{2\eta(\eta^2 - m^2) + \eta - m^2 \pm [(\eta - m^2)^2 + 4m^2(\eta^2 - \eta)(\eta^2 - m^2)]^{1/2}\}/4(\eta^2 + m^2). \quad (21)$$

At a fixed frequency and direction of the pump wave, the Δ obtained in this manner determines the synchronous pitch. Keeping the other parameters fixed, we can determine the synchronous angle or frequency from the obtained value of Δ.

Generally speaking, the synchronism condition is reached in this case not on the boundary of the region of selective reflection.

The condition for reaching synchronism on this boundary is that the parameter Δ coincide with the values corresponding to the boundaries of the region of selective reflection.[3,10] The dependence of the boundary values Δ_e on the direction of propagation of the harmonic $\mathbf{k}_0$ (see Fig. 3) is the following. The three regions of the polarization-dependent light reflection are bounded respectively by the values Δ_e^p: $-(1 + (1 + 8m^2)^{1/2})/2$, 0 and $((1 + 8m^2)^{1/2} - 1)/2$, 1. The expressions for the boundaries of the region of reflection of light of arbitrary polarization Δ_e^T depend on the beam incidence angle. In the angle range containing the normal incidence $32° \leq \theta \leq 90°$ $((2 + \sqrt{13} \geq m^2 \geq 0)/18)$ the limits of Δ_e^T are zero and $((1 + 8m^2)^{1/2} - 1)/2$, i.e., the regions of

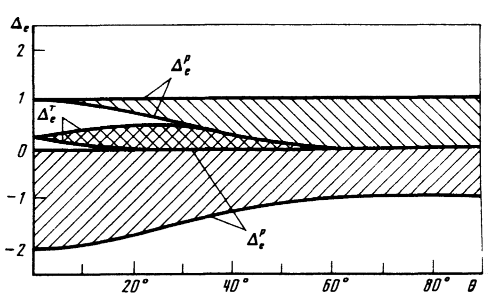

FIG. 3. Angular dependences of the boundaries Δ_e^T and Δ_e^p of the regions of selective reflection (the region of reflection of light of arbitrary polarization is shown cross hatched).

selective and nonselective reflection of the polarizations are in contact with each other. For $25° \leqslant \theta \leqslant 32°$ ($\frac{1}{2} \geqslant m^2 \geqslant (2+\sqrt{13})/2)\Delta_e^T$ they are equal to zero and $((1+8m^2)^{1/2}-1)/2$, i.e., one of the regions of the selective reflection is split off, and for $0° \leqslant \theta \leqslant 25°$ ($1 \geqslant m^2 \geqslant \frac{1}{2}$) both regions of selective reflection are split off from the region of the nonselective reflection of the polarizations, the boundaries of which Δ_e^T are now

$$[m-(5m^2-4m^4-1)^{1/2}]/4m \text{ and } [m+(5m^2-4m^4-1)^{1/2}]/4m.$$

From the solution of the system (18) it follows that, just as in the case of normal propagation of the pump wave, when synchronism is reached near the boundary of the region of selective reflection, i.e., when relation (20) is satisfied for values of Δ close to those shown in Fig. 3, the efficiency of harmonic generation increases sharply, and its amplitude can be proportional to the square of the sample thickness.

The general expressions for the solutions of the system (18) become simpler in case of synchronism near the boundary of the regions of selective reflection. In particular, if the synchronism takes place exactly on the boundary $\Delta = \Delta_e^b$, the polarization characteristics of the generated waves, for sufficiently thick samples, coincide with the characteristics of the corresponding eigensolutions and are described by the polarization vectors

$$\mathbf{n}_q = \boldsymbol{\sigma} \cos\psi + i\boldsymbol{\pi}_q \sin\psi, \qquad \operatorname{tg}\psi = \sin\theta \frac{\Delta+m}{\Delta-m}, \quad q=0,1; \tag{22}$$

where $\boldsymbol{\sigma}$ and $\boldsymbol{\pi}_q$ are linear-polarization unit vectors perpendicular to the $(\mathbf{k}_0, \mathbf{k}_1)$ plane and lying in the $(\mathbf{k}_0, \mathbf{k}_1)$, plane respectively.

The maxima of the intensity of the harmonic are reached when the synchronism condition (19) is satisfied almost on the boundary of the region of selective reflection for discrete values of $\Delta = \Delta_s$, defined by the relation

$$L^2(\mathbf{k}_{0+}-\mathbf{k}_{0-})^2 = A_e(\Delta_s - \Delta_e) = (2\pi s)^2,$$

where s is an integer, $\mathbf{k}_{0\pm}$ are the wave vectors of those eigensolutions of the linear optical problem which coincide at $\Delta = \Delta_e$, and L is the thickness of the cholesteric layer. Without writing out the expressions for $\mathbf{k}_{0\pm}$ in the general case (see Ref. 9), we present the expressions for the values of Δ_s near the boundary $\Delta_e = 0$:

$$\Delta_s = (2\pi s)^2(1-m^2)(2m^2-1)/2m^2(\delta\varkappa_s L)^2.$$

The frequency of the harmonic coincides with the boundary $\Delta_e = 0$ if the following relation holds between the dielectric anisotropy, the dispersion, and the propagation direction of the pump wave $\mathbf{k}^\sigma(\omega)$:

$$(\bar{\varepsilon}_3 - \bar{\varepsilon}_1)/\bar{\varepsilon}_1 = \delta \cos^2\theta/(2+\sin^2\theta).$$

If it is not assumed that the distance of the synchronous-generation frequency ω^s, defined with the aid of reflection (21), from the boundary of the region of selective reflection is small, the harmonic-generation efficiency falls off in oscillatory fashion and tends to the known expressions far from the region of selective reflection.[8]

CONCLUSION

The presented analysis of the nonlinear-optical conversion of light frequency in a cholesteric under conditions when the harmonic is close to the region of selective reflection demonstrates that light diffraction in a cholesteric can influence substantially the characteristics of the nonlinear processes. In addition to the known difference between the synchronism conditions of periodic and homogeneous media,[11] a substantial increase of the efficiency of the nonlinear process can appear. Although we have analyzed above third-harmonic generation, it is clear, in view of the rather general nature of the phenomenon, that a similar increase of the efficiency takes place also for all periodic media and for other nonlinear processes, e.g., second-harmonic generation.

We indicate also that a similar increase in the intensity of Cherenkov radiation was observed in periodic media at a frequency equal to the boundary of the selective-reflection region.[12,13] As for cholesteric liquid crystals and chiral smectic crystals, they are the most convenient objects for the observation of the growth, revealed here, of the efficiency of nonlinear conversion, since they make it possible to vary relatively simply the parameters that are of importance for the problem in question, namely the period of the structure and the anisotropy of the dielectric properties.

When speaking of the quantitative aspect of the expected experimental increase in the intensity, an increase, say, of the intensity of the harmonic by a factor 10–100 imposes rather reasonable requirements on the experimental conditions and can be reached at the following values of the parameters: pump line width and wavelength respectively $\Delta\omega/\omega \sim 10^{-5}$, $\lambda \sim 10^4$ Å and cholesteric parameters $L \sim 0.01$ cm and $\delta \approx 0.1$.

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APPENDIX

We write down explict expressions for the Fourier expansion of the polarization in a cholesteric, needed for the solution of Eqs. (3) and (18). We consider the case of propagation of a pump wave $\mathscr{E}_\pm(z,\omega)$ along the optical axis. We represent the eigenwaves in the cholesteric in the form[1]

$$\begin{aligned}\mathscr{E}_\pm(z,\omega) &= \mathscr{E}_\omega^\pm[\hat{n}_+(1-f_\omega^\pm)\exp\{i(k_\omega^\pm+\tau/2)z\} \\ &+ \hat{n}_-(1+f_\omega^\pm)\exp\{i(k^\pm-\tau/2)z\}][2(1+|f_\omega^\pm|^2)]^{-1/2}, \\ f_\omega^\pm &= \tau k_\omega^\pm[(k_\omega^\pm)^2+\varkappa_\omega^2(\delta_\omega-1)+\tau^2/4]^{-1}.\end{aligned} \tag{A1}$$

Using the periodicity of the properties of the NLC, we can expand the nonlinear-susceptibility third-rank tensor $\chi^{(3)}(z,3\omega)$ in a Fourier series. With the aid of expression (A1) for the eigenwaves of the cholesteric, we represent the vector of the nonlinear polarization in the form

$$P_{(z,3\omega)}^{NL} = -\frac{\bar{\varepsilon}_3}{4\pi}\sum_{slmn}(\hat{n}_+P_{lmn}^{+s}e^{i\tau z/2}+\hat{n}_-P_{lmn}^{-s}e^{-i\tau z/2})\exp\{i(k_\omega^l+k_\omega^m+k_\omega^n+s\tau)z\} \cdot \tag{A2}$$

For the case when the cholesteric can be regarded locally as equivalent to a nematic, i.e., as a centrosymmetric and axisymmetric medium, the only nonzero amplitudes in the expansion (A2) are $P^{\pm}_{lmn}$ for $s=0$:

$$P_{lmn}=\varepsilon_{\omega}{}^{l}\varepsilon_{\omega}{}^{m}\varepsilon_{\omega}{}^{n}[c_{11}\mp if_{\omega}{}^{l}c_{21}+(-c_{12}\pm f_{\omega}{}^{l})f_{\omega}{}^{m}f_{\omega}{}^{n}][2(1+|f_{\omega}{}^{l}|^{2})\times(1+|f_{\omega}{}^{m}|^{2})(1+|f_{\omega}{}^{n}|^{2})]^{-1/2}, \tag{A3}$$

where c_{ik} $(i,j=1,2)$ are the z-independent components of the nematic local nonlinear-susceptibility tensor $\chi^{T(3)}$:

$$c_{11}=\chi^{T(3)}_{1111}; \quad c_{22}=\chi^{T(3)}_{2222}=\chi^{T(3)}_{3333}=3\chi^{T(3)}_{2233}=3\chi^{T(3)}_{3322},$$

$$c_{12}=3\chi^{T(3)}_{1122}=3\chi^{T(3)}_{1133}, \quad c_{21}=3\chi^{T(3)}_{2211}=3\chi^{T(3)}_{3311}.$$

[1]J. W. Shelton and Y. R. Shen, Phys. Rev. A5, 1867 (1972).
[2]S. M. Arakelyan, G. A. Lyakhov and Yu. S. Chilingaryan, Usp. Fiz. Nauk **131**, 3 (1980) [Sov. Phys. Usp. **23**, 245 (1980)].
[3]V. A. Belyakov, V. E. Dmitrenko, and V. P. Orlov, Usp. Fiz. Nauk **127**, 221 (1979) [Sov. Phys. Usp. **22**, 63 (1979)].
[4]C. Elachi, Proc. IEEE **64**, 1666 (1976).
[5]V. A. Belyakov and N. V. Shipov, Phys. Lett. **A86**, 94 (1981).
[6]V. G. Kamenskiĭ and E. I. Katz, Opt. Spektrosk. **45**, 1106 (1978).
[7]P. P. Bey and H. Rabin, Phys. Rev. **162**, 794 (1967).
[8]N. V. Shipov and V. A. Belyakov, Zh. Tekh. Fiz. **50**, 205 (1980) [Sov. Phys. Tech. Phys. **25**, 125 (1980)].
[9]V. A. Belyakov and V. E. Dmitrenko, Fiz. Tverd. Tela (Leningrad) **15**, 2724 (1973) [Sov. Phys. Solid State **15**, 1811 (1974)].
[10]V. E. Dmitrenko, Candidate's dissertation, Crystallography Inst., Moscow, 1978.
[11]A. A. Maier, A. P. Sukhorukov, and R. N. Kuz'min, Zh. Eksp. Teor. Fiz. **77**, 1282 (1979) [Sov. Phys. JETP **50**, 645 (1979)].
[12]E. I. Katz, Zh. Eksp. Teor. Fiz. **61**, 1686 (1971) [Sov. Phys. JETP **34**, 899 (1972)].
[13]V. A. Belyakov, V. E. Dmitrenko, and V. P. Orlov, Pis'ma Zh. Tekh. Fiz. **1**, 978 (1975) [Sov. Tech. Phys. Lett. **1**, 422 (1975)].

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