# DOUBLING OF LASER RADIATION FREQUENCY UNDER NONSTATIONARY CONDITIONS

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Results are presented of an experimental and theoretical investigation of optical harmonic generation by picosecond laser pulses. A nonstationary theory of harmonic generation is developed, in which space and time wave modulation are taken into account. The analysis is based on the quasi-optics equations, in which the dispersion properties of the nonlinear crystals are taken into account in the first approximation of dispersion theory. The analysis is performed for frequency-modulated pulses. It is shown that for nonstationary generation conditions the spectral and angular distributions of the harmonic are interrelated and the space-time structure of the harmonic may appreciably differ from those of the laser beam. Beam focusing under nonstationary conditions is analyzed and the condition for optimal focusing is derived. An expression for the maximal energy of the second harmonic is obtained. In the experimental investigation, the main attention is directed to the spectral and angular distributions. These have been obtained in quasistatic (KDP crystal) as well as nonstationary ($LiNbO_3$ crystal) frequency doubling conditions. Variation of the shape and width of the harmonic spectrum are determined by varying the divergence of the laser beam. The experimental results are in agreement with the theoretical results.

IN connection with the progress of laser physics in the field of generation of ultrashort light pulses, considerable attention is now paid by investigators to the nonstationary optical phenomena. In the field of very short, picosecond laser pulses, the character of the nonlinear optical processes is in many respects different from that in the field of pulses of microsecond and nanosecond duration. This circumstance is connected with the fact that the duration of the ultrashort pulses ($\tau \sim 10^{-12}$ sec) is comparable with the time of relaxation of the nonlinearities of media and the times of group delay of the interacting light waves. In the general case, both these effects become manifest, naturally, simultaneously. However, in many processes, such as the generation of harmonics and parametric processes, the only important factor in the finite character of the time of the group delay of the waves[1,2], which causes the nonstationarity behavior of the wave.

For the nonstationary case, a theoretical analysis of the generation of a second optical harmonic by ultrashort pulses and broad-spectrum emission has been carried out for plane waves in[1,3-6]. The experimental investigation of the generation of a harmonic in a nonstationary regime, corresponding to such a model, was recently carried out by Shapiro[1)] [7], who investigated the dependence of the duration of the ultrashort harmonic pulses on the length of the nonlinear $LiNbO_3$ crystal. Even earlier, Kovrigin[8] observed changes in the angle structure of the second harmonic, excited in a KDP crystal by a broad spectrum (~80 Å), in comparison with that obtained for a narrow spectrum.

By now, the analysis of the generation of optical harmonics has been carried out separately for spatially-bounded beams (see, for example,[9,10]) and for pulses (see[1]). However, for ultrashort pulses, the spatial boundedness of the beams can be of fundamental significance. As we shall show below, in this case the effects connected with the spatial and temporal modulations of the waves can greatly influence one another.

In this article, we investigate theoretically the generation of second harmonics by spatially-bounded beams (diverging and focused) with broad frequency spectra (ultrashort pulses and pulses with frequency modulation). Some of the results of the calculations are compared with data of an experiment performed by the authors on second-harmonic generation in $LiNbO_3$ and KDP crystals with beams from a neodymium laser with synchronized modes. Principal attention was paid here to the analysis in the nonstationary regime of the angle structure and the frequency spectrum of the second harmonic.

## 1. THEORY OF GENERATION OF OPTICAL HARMONICS BY SPATIALLY-BOUNDED BEAMS WITH BROAD FREQUENCY SPECTRA

The process of doubling the frequency of laser radiation with simultaneous account of the spatial and temporal modulation at small harmonic-conversion coefficients is described by the following equations[1]:

$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_2}\frac{\partial}{\partial t}+\beta\frac{\partial}{\partial x}+i\frac{1}{2k_2}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)\right\}A_2=-i\sigma A_1^2e^{-i\Delta z}, \quad (1a)$$

$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_1}\frac{\partial}{\partial t}+i\frac{1}{2k_1}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)\right\}A_1=0. \quad (1b)$$

The notation here is standard: the index 1 pertains to the fundamental radiation, index 2 to the second harmonic, $A_n$ are the complex amplitudes of the wave, $u_n$ the group velocities, $k_n$ the wave numbers, $\Delta = 2k_1 - k_2$ is the deviation of the wave numbers, $\beta$

[1)] We note that in [7] the synchronism angle was chosen to be $\theta_s = 90°$ by varying the crystal temperature. In this case there are no effects connected with the finite dimensions and the angle divergence of the laser beam.

is the birefringence angle[2)], $\sigma$ the coefficient of nonlinear coupling, the z axis is directed along the normal to the boundary between the linear and nonlinear media, and the x axis lies in the plane of the birefringence.

Equation (1) constitute a system of equations of wave optics, in which the dispersion properties of the medium are taken into account in first approximation of dispersion theory. These equations will be solved with the following conditions on the interface z = L

$$A_1(t, x, y, z = L) = A_{10}(t, x, y, L), \quad A_2(t, x, y, z = L) = 0. \quad (2)$$

Although the solution of Eqs. (1) can be obtained directly, it will be more convenient in the subsequent analysis to go over immediately in (1) to the frequency-wave spectrum by means of the transformation

$$S_n(\Omega, \varkappa, z) = \frac{1}{(2\pi)^3}\iiint_{-\infty}^{\infty} A_n(t, x, y, z)\, e^{-i(\Omega t - \varkappa\rho)}\, dt\, d\rho, \quad (3)$$

where $\kappa\cdot\rho = k_x x + k_y y$ and $d\rho = dxdy$.

For the second harmonic, such a spectrum is determined by the expression

$$S_2(\Omega,\varkappa,l) = -i\sigma\exp\{-i\Delta L - il\psi_2(\Omega,\varkappa)\}\int_{-\infty}^{\infty}\dots\int S_{10}(\Omega_1,\varkappa_1,L)$$
$$\times S_{10}(\Omega_2,\varkappa_2,L)\, e^{-il\psi/2}\frac{\sin(\frac{1}{2}l\psi)}{\frac{1}{2}\psi}\,\delta(\Omega-\Omega_1-\Omega_2)$$
$$\times\,\delta(\varkappa-\varkappa_1-\varkappa_2)\, d\Omega_1\, d\Omega_2\, d\varkappa_1\, d\varkappa_2, \quad (4)$$

where $l$ is the length of the nonlinear crystal, $S_{10}(\Omega, \kappa, L)$ is the spectrum of the fundamental radiation, $\Omega$ is the deviation from the central frequency $\omega_1$,

$$\psi = \Delta + \psi_1(\Omega_1, \varkappa_1) + \psi_1(\Omega_2, \varkappa_2) - \psi_2(\Omega, \varkappa),$$

$$\psi_1(\Omega_n, \varkappa_n) = \frac{\Omega_n}{u_1} - \frac{1}{2k_1}\varkappa_n^2, \quad \psi_2(\Omega,\varkappa) = \frac{\Omega}{u_2} - \beta k_x - \frac{1}{2k_2}\varkappa^2.$$

From an analysis of (4) it is easy to obtain the condition for the maximal contribution of the spectral components of the fundamental radiation to the harmonic; this will obviously be the condition of vector synchronism (compare with[1,10]):

$$\mathbf{k}_1(\omega_1 + \Omega_1) + \mathbf{k}_1(\omega_1 + \Omega_2) = \mathbf{k}_2(2\omega_1 + \Omega). \quad (5)$$

Analytic results can be obtained from (4) only for concrete models of the laser radiation. For the case of plane waves and monochromatic beams, expression (4) leads to the distributions obtained in[1] for the spectra of the harmonic.

In the present paper we considered harmonic excitation by a Gaussian beam, the amplitude of which in the transverse cross section at z = 0 is given by[3)]

$$A_1(t,\rho) = A_0\exp\left\{-\frac{\rho^2}{a^2} - \left(\frac{1}{\tau^2} + i\gamma\right)t^2\right\}, \quad A_0 = \left(\frac{16}{ca^2\tau}W_1\sqrt{\frac{2}{\pi}}\right)^{1/2}, \quad (6)$$

where $W_1$ is the total energy of the beam, and the parameter $\gamma$ characterizes the frequency modulation.

If a Gaussian beam passes through a lens, say a spherical one, located in the Fresnel zone of the beam, then the amplitude of the beam at z = 0 is described by the expression

$$A_{10}(t,\rho) = A_0\exp\left\{-\left(\frac{1}{a^2} + i\frac{k}{2R_1}\right)\rho^2 - \left(\frac{1}{\tau^2} + i\gamma\right)t^2\right\}. \quad (7)$$

Here k is the wave number in the linear medium, and $R_1$ is the focal length of the lens (with the aid of $R_1$ it is possible to take into account also the presence of a definite angular divergence of the laser radiation). Solving Eq. (1b) and taking (7) into consideration, we obtain an expression for the spectrum of the laser radiation at a distance z from the lens:

$$S_1(\Omega,\varkappa,z) = S_{10}(\Omega,\varkappa)\exp\left\{i\left[\frac{1}{2k}\varkappa^2 - \frac{\Omega}{u}\right]z\right\}, \quad (8)$$

where $S_{10}(\Omega, \kappa)$ is the Fourier transform of the expression (7).

Finally, substitution of (8) in (4) leads after straightforward but cumbersome calculations to the second-harmonic spectrum for the considered model of the laser radiation:

$$S_2(\Omega,\varkappa,l) = -i\sigma A_0^2\frac{\tau a^2}{8(2\pi)^{3/2}}\exp\left\{-\frac{\tau^2\Omega^2}{8(1+i\gamma\tau^2)} - \frac{a^2\varkappa^2}{8(1-iD/R)} - i\varphi\right\}$$
$$\times\frac{1}{(1-iD/R)\sqrt{1+i\gamma\tau^2}}\int_{nL}^{nL+l}\frac{\exp\{-iz[\Delta+\beta k_x-\nu\Omega]\}}{1-iz(1/D-i/R)}dz. \quad (9)$$

Here $D = (1/2)k_1a^2$ is the diffraction length of the beam, $R = nR_1$, n is the refractive index in the synchronism direction, $\nu = u_2^{-1} - u_1^{-1}$ is the detuning of the group velocities, and the phase $\varphi$ is equal to

$$\varphi = \left[\frac{\Omega}{u_2} - \beta k_x - \frac{1}{2k_2}\varkappa^2\right]L - \frac{L}{4k}\varkappa^2 + \frac{L}{u}\Omega + \Delta l.$$

Using (9), let us consider different characteristics of the second-harmonic radiation.

1. Spectral characteristics of the harmonic. The distribution of the spectral density over the frequencies (usually called the spectral distribution) and the distribution of the spectral density over the wave numbers $k_x$, and $k_y$ (or the angular distribution) of the second harmonic, registered respectively by a spectrograph and by a photographic plate placed at the focus of a gathering lens, are determined by the formulas

$$I_2(\Omega,l) = 4\pi^2\iint|S_2(\Omega,\varkappa,l)|^2 d\varkappa,$$

$$I_2(\varkappa,l) = 2\pi\int|S_2(\Omega,\varkappa,l)|^2 d\Omega. \quad (10)$$

For the case $S_2(\Omega, \kappa, l)$ in the form (9), the distributions $I_2(\Omega, l)$ and $I_2(\kappa, l)$ are given by the expressions

$$I_2(\Omega,l) = \frac{K\tau}{2\sqrt{\pi}[1+\gamma^2\tau^4]}\exp\left\{-\frac{\tau^2\Omega^2}{4(1+\gamma^2\tau^4)}\right\}F_\nu(\Omega,\beta,l), \quad (11)$$

$$I_2(\varkappa,l) = \frac{Ka^2}{4\pi(1+D^2/R^2)}\exp\left\{-\frac{a^2\varkappa^2}{4(1+D^2/R^2)}\right\}F_\beta(k_x,\nu,l), \quad (12)$$

where

$$K = 8\sigma^2W_1^2/\sqrt{\pi}ca^2\tau, \quad (13a)$$

$$F_\nu(\Omega,\beta,l) = \iint_{nL}^{nL+l}\Phi(z_2,z_1)$$
$$\times\exp\left\{i\nu\Omega(z_2-z_1) - \frac{\beta^2}{a^2}(1+D^2/R^2)(z_2-z_1)^2\right\}dz_1\,dz_2. \quad (13b)$$

$$F_\beta(k_x,\nu,l) = \iint_{nL}^{nL+l}\Phi(z_2,z_1)$$

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2) In the present paper, the investigation is limited to an interaction of the type oo → e (o – ordinary, e – extraordinary waves).

3) A Gaussian distribution of intensity in the cross section of the beam is possessed not only by lasers with spherical and semispherical resonators [11], but, as shown by the experimental data, also by solid-state lasers with flat mirrors, owing to the inhomogeneities of the crystals (see, for example, [12]).

$$\times \exp\left\{-i\beta k_x(z_2-z_1)-\frac{\nu^2}{\tau^2}(1+\gamma^2\tau^4)(z_2-z_1)^2\right\}dz_1\,dz_2, \quad (13c)$$

$$\Phi(z_2, z_1) = \{[1-iz_2(1/D-i/R)][1+iz_1(1/D+i/R)]\}^{-1} \times\exp\{-i\Delta(z_2-z_1)\}. \quad (13d)$$

It is seen from (12) that the angular distribution of the harmonic along the y axis is determined only by the distribution of the fundamental radiation along the same axis. As to the angular distribution of the harmonic with respect to x, it can depend strongly on the properties of the nonlinear crystal, $\beta$ and $\nu$, and on the temporal characteristics of the main beam $(\tau, \gamma)$. Similarly, the spectral distribution of the harmonic (11) can be determined to a considerable degree by the parameters $\nu$ and $\beta$ and the spatial characteristics of the fundamental radiation (a, D, R). From a comparison of (11) and (12) it is also seen that, in accordance with the space-time analogy in nonlinear optics[1], the angular distribution of the harmonic with respect to x can be obtained from the spectral distribution with the aid of the substitution[4)]

$$\Omega\to k_x,\quad \tau\to a,\quad \gamma\tau^2\to D/R\quad (\gamma\to k/2R_1),\quad \nu\to-\beta. \quad (14)$$

Taking this circumstance into account, let us examine in detail one type of distribution, namely the spectral distribution.

Spectral distribution. The form of the distributions (11) depends on the relation between the length of the crystal $l$, the aperture and quasistatic characteristic lengths $l_a$ and $l_q$, respectively, and the values of D and R. Let us examine some of them, bearing in mind mainly a comparison of the results of the calculations with the experimental results.

Let us consider first the case of harmonic excitation by a plane-parallel beam $(R\to\infty)$, when the crystal is in the Fresnel zone of the laser radiation. $|D \gg (nL = l)|$. In general form, the function $F_\nu$ (13b) is expressed here in terms of probability integrals with complex arguments. If the crystal length $l$ is smaller than the aperture length $l_a = a/\beta$, then $F_\nu$ assumes the known form for plane waves:

$$F_\nu(\Omega)=\frac{\sin^2\{(\nu\Omega-\Delta)l/2]}{\{(\nu\Omega-\Delta)/2]^2}. \quad (15)$$

From (11) and (15) it follows that at a length $l$ smaller than the quasistatic length $l_q = \tau/|\nu|\sqrt{1+\gamma^2\tau^4}$, we get $E_\nu(\Omega) = l^2$ and the spectral distribution of the harmonic is Gaussian; this is the so-called quasistatic or quasistationary regime. In the nonstationary regime $(l > l_q)$, i.e., when the width of the fundamental-radiation spectrum is

$$\Delta\omega_1 = 2{,}34\frac{\sqrt{1+\gamma^2\tau^4}}{\tau} > \frac{2\pi}{|\nu|l},$$

the form of the spectrum of the harmonic is determined essentially by the function (15), and in the case of a detuning $\Delta\neq 0$ the maxima of the same order in the wings of the spectral distribution of the harmonic have different values. In the general case, the width of the spectrum of the harmonic can be determined from the expression

$$\Delta\omega_2 = \Delta\omega_{2,k}/\sqrt{1+0{,}357(l/l_g)^2}, \quad (16)$$

where $\Delta\omega_{2,k} = 3.32\sqrt{1+\gamma^2\tau^4}/\tau$ is the width of the spectrum in the quasistatic regime. In the nonstationary regime $\Delta\omega_2 = 5.56/|\nu|l$.

If the crystal length $l$ is greater than the aperture length, then the spectral distribution begins to depend on the value of $l_a$. When $l \gg l_a$, the function $F_\nu$ is given by

$$F_\nu = \sqrt{\pi}ll_a\exp\{-{}^1/_4(\nu\Omega-\Delta)^2l_a^2\}. \quad (17)$$

The width of the spectrum of the harmonic is determined as before by expression (16), in which the quantity $0.58l$ should be replaced by $l_a$. Thus, owing to the influence of the aperture effect on the generation process, the spectral distribution of the harmonic in the nonstationary regime becomes smoothed out, and the width of the spectrum $\Delta\omega_2$ increases. By changing for a given crystal the value of the aperture length $l_a$, in other words, by changing the dimension of the beam a, we can vary $\Delta\omega_2$. As expected, the dispersion of the medium ceases to influence the spectrum of the harmonic when the effects connected with the finite dimension of the beam become manifest earlier than the temporal effects $(l_a < l_q)$, and consequently the process of harmonic generation becomes quasistatic. The presence in (17) of a detuning $\Delta\neq 0$ leads to a change in the value of the average frequency of the harmonic, by an amount

$$\Omega_0 = \Delta/\nu[1+(l_q/l_a)^2].$$

Let us examine now the spectra of the harmonic excited by focused beams. The case of focusing of the beam on the front face of the crystal, when the conditions $l \ll R$ and $D \gg (nL + l)$ are satisfied, has in fact just been analyzed. When the beam is focused in the center of the crystal, an analysis of the spectral distribution of the harmonic, if the length of the crystal is smaller than the aperture length $l_a$, is identical with the analysis of the angular distribution for a monochromatic beam[10].

Let us stop to discuss in greater detail the example of harmonic generation by defocused beams. In the case of strong defocusing of the beam $(|R| \ll D)$ and $\beta\neq 0$, the function $F_\nu$ (13b) is analogous to the expression (17), where the role of the aperture length is played by the quantity $l_a' = aR/\beta D$. If $\beta = 0$, then the function $F_\nu$ is given by

$$F_\nu(\Omega) = |R|^2\{[\mathrm{ci}(\xi)-\mathrm{ci}(\eta)]^2+[\mathrm{si}(\xi)-\mathrm{si}(\eta)]^2\}, \quad (17')$$

where

$$\xi=\eta+l|\nu\Omega|,\quad \eta=(nL+|R|)|\nu\Omega|.$$

An analysis shows that in this case the spectral distribution of the harmonic can be close to a Gaussian distribution with weak modulation.

Angular distribution. As already noted, the angular distribution of the harmonic (12) can be obtained from the spectral distribution (11) by using the substitution (14). Thus, for example, the substitution of (14) in expressions (15) and (16) gives the form of the function $F_\beta(k_x)$ (13c) at $D \gg (nL + l)$ and $R\to\infty$. It follows therefore that the angular spectrum of the harmonic is discrete-continuous in the quasistatic generation

4) We note at the same time that the quantity D itself does not have a temporal analog here, since different approximations have been used to describe the spatial and temporal modulation of the wave (see [1]).

regime, $l < l_q$ (such a structure of the frequency spectrum takes place in the nonstationary regime when $l < l_a$), and becomes continuous when $l \gg l_q$. Thus, the character of the angular distribution of the harmonic and its width now depends strongly on the dispersion properties of the crystal and on the spectral width of the fundamental radiation. Consequently, it becomes possible to vary the angular divergence of the harmonic.

2. Space-time characteristics of the harmonic. No less important characteristics of the harmonic are the spatial dimensions of the beam and the pulse duration. Although these parameters are always connected with the spectral distribution, in the case of harmonic generation in the nonstationary regime, owing to the fact that, as shown above, they influence each other, the space-time structure of the beam becomes sufficiently complicated and calls for a separate consideration. Let us illustrate the foregoing using the simplest problem of harmonic generation by a plane-parallel beam ($R \to \infty$, $D \to \infty$). In this case the complex amplitude of the second harmonic is determined by the expression

$$A_2(t,x,y,l) = -i\sigma A_0^2 \int_0^l \exp\left\{-2\left[\frac{1}{\tau^2}+i\gamma\right] \times (t-T+\nu\xi)^2 - 2\frac{y^2+(x-\beta l+\beta\xi)^2}{a^2}\right\} d\xi, \qquad (18)$$

where $T = L/u + l/u_2 - \nu nL$. When $l \gg l_q$ and (or) when $l \gg l_a$, Eq. (18) takes the form

$$A_2(t,x,y,l) = -i\sigma A_0^2 \exp\{-2[\beta t - \nu x + \beta x_0]^2/(d - ib\gamma)\}. \qquad (19)$$

Here

$$b = \beta^2(\tau^{-4}+\gamma^2)^{-1}, \quad d = a^2\nu^2 + b\tau^{-2},$$

$$x_0 = \frac{L}{u} + \frac{l}{u_2} - \nu(nL + l).$$

Thus, when the fundamental-radiation beam is frequency modulated ($\gamma \neq 0$) the harmonic radiation has both frequency and (if $\beta \neq 0$) spatial modulation of the phase (an angular divergence), and the index of the modulation depends in a complicated manner on the values of the quantities $\gamma$, $\beta$, $\tau$, a, and $\nu$. At a given instant of time t, the maximum value of the harmonic intensity takes place at the point $x = \beta(t + x_0)/\nu$.

From (19) we can readily determine the duration of the pulse $T_2$ and the beam width $X_2$ of the harmonic. In the case of beams without frequency modulation ($\gamma = 0$), $T_2$ is given by (compare with (17) with $0.57l \to l_a$)

$$T_2 = T_{2,q}\sqrt{1 + (l_a/l_q')^2}, \qquad (20)$$

where $l_q' = \tau/|\nu|$.

Consequently, the harmonic pulse duration in the nonstationary regime is always larger than in the quasistatic regime $T_{2,q} = 0.83\tau$. For beams with frequency modulation, $T_2$ can be either larger or smaller than the quasistatic value. This is determined by the ratio of the lengths $l_a$, $l_q$, and $l_q'$. For example, if the width of the spectrum of the fundamental radiation is determined principally by the frequency modulation, then when $l_q' > l_a > l_q = \tau/|\nu|\sqrt{1+\gamma^2\tau^4}$ we get

$$T_2 = T_{2,q}\sqrt{(l_a/l_q')^2 + (l_q'/\gamma\tau^2 l_a)^2}. \qquad (21)$$

We recall that in the nonstationary regime the width of the spectrum of the harmonic is always narrower in our problem than in the quasistatic regime.

The foregoing results can be extended also to the harmonic beam widths.

3. Energy of harmonic. Conditions for optimal focusing of pulses with a broad spectrum. The expression for the energy $W_2$ of the second harmonic can be obtained by integrating (11) or (12); we then have

$$W_2 = K \int\int_{nL}^{nL+H} \Phi(z_2,z_1)\exp\left\{-\left[l_q^{-2} + \left(1+\frac{D^2}{R^2}\right)l_a^{-2}\right](z_2-z_1)^2\right\} dz_2\,dz_1. \qquad (22)$$

In the limit as $l_q \to \infty$ ($\tau \to \infty$) we obtain a formula for the power of the harmonic excited by a monochromatic beam. Thus, both expressions turn out to be analogous, the only difference being that the argument of the exponential in (22) includes also the quasistatic length $l_q$; these expressions can be reduced to a single form by introducing in (22) the effective "birefringence angle":

$$\beta_{\text{eff}} = \beta\sqrt{1 + l_a^2/l_q^2(1 + D^2/R^2)}. \qquad (23)$$

An analysis of the energy of the harmonic $W_2$ under different generation conditions can be readily carried out by following[10]. Let us emphasize the features of beam focusing with a broad frequency spectrum using as an example a harmonic excited by a beam focused at the center of the crystal ($L = R - l/2n$). Since the conditions for optimal focusing for monochromatic beams do not depend on the birefringence angle $\beta$, in the focusing of nonmonochromatic they will likewise not depend on $\beta_{\text{eff}}$. For the problem under consideration, the optimal radius of the focusing lens is $R_{10} = 0.42na\sqrt{k_1 l}$. In the nonstationary generation regime ($l_q < l$), the maximum energy of the harmonic, obtained at optimal focusing, now depends not only on the length of the crystal, but also on the value of the characteristic length $l_a$ and $l_q$ ($R < D$).

$$W_{2,\,max} \approx 6.45\,\sigma^2 W_1^2 (k_1 l)^{1/2}/\beta\tau\sqrt{1 + (l_a R/l_q D)^2}. \qquad (24)$$

If the quasistatic length $l_q$ is much smaller than the aperture length $l_a$ (for example, in the generation of harmonics in the direction of the synchronism angle $\theta_s = 90°$, $\beta = 0$), then (24) takes the form

$$W_{2,\,max} = 6.45\,\sigma^2 W_1^2 D (k_1 l)^{1/2}/aR|\nu|\sqrt{1+\gamma^2\tau^4}. \qquad (25)$$

Thus, in the case under consideration, the maximum energy of the harmonic is influenced by the dimension of the beam a and its frequency modulation. It should be noted that, at an equal width of the frequency spectrum, frequency-modulated beams produce a smaller energy value $W_{2,max}$ ($W_{2,max}$ does not depend at all on the pulsed duration $\tau$ when $\gamma = 0$).

## 2. EXPERIMENTAL INVESTIGATION OF HARMONIC GENERATION BY ULTRASHORT PULSES

1. Parameters of the apparatus. In the experiment we used as the source of the second-harmonic excitation the radiation from a neodymium-glass laser operating with self-synchronized modes. The active element (the laser rod) had end faces inclined at the Brewster angle. The resonator length was 130 cm. The resonator mirrors were coated on wedge-like substrates. The reflection coefficient of the "dead"

mirror was 99%, and that of the output mirror 53%. The initial transmission of the saturable filter was 50%.

The laser emission had the following characteristics: total train energy ~0.1 J, number of pulses in the train 10—15, pulse duration $(3-4)\times 10^{-12}$ sec; the power density reached 30 GW/cm$^2$, the beam divergence at half intensity level ~1′, and the beam diameter 3 mm.

The duration of the ultrashort pulses was estimated by the two-photon procedure[14], the criterion for the synchronization of the laser modes being the ratio R of the maximum intensity to the background. The measured values $R \approx 1.8-2.3$ (see Fig. 1) lie between the theoretical values for unsynchronized modes, $R = 1.5$, and for fully synchronized modes, $R = 3$[13]. There was good agreement between the laser-emission spectrum determined from measurements on the pulse duration and the width of the spectrum measured with the aid of a spectrograph. Consequently, in our case the laser radiation had no significant frequency modulation.

The second-harmonic generation was investigated in KDP crystals 2.5 cm long and $LiNbO_3$ crystals 1 cm long. This set of crystals has made it possible, without retuning the laser generator, to compare the different excitation regimes of the harmonic (quasistatic $l < l_q$). Indeed, for pulses of duration $3\times 10^{-12}$ sec, the quasistatic lengths for the KDP and $LiNbO_3$ crystals were respectively 25 cm and 0.5 cm. The generation of the harmonic was effected at small conversion coefficients (less than 5%).

2. Experimental results. Discussion. The observed experimental spectra of the almost plane-parallel beam of second harmonic, but out by the spectrograph slit (0.03 mm) from the generated weakly diverging beam, are shown in Fig. 2. Figure 3 shows the corresponding photometric curves. The spectra of the harmonic emerging from the KDP is continuous, its width $\Delta\lambda_2 = 6$ Å corresponds to the quasistatic conversion regime. The spectral distribution of the harmonic excited in $LiNbO_3$ is highly uneven. The width of the central peak between the minima is 3 Å, the width of the side peaks to the left of the maximum is $\Delta\lambda_2(e) = 1.8$ A, and on the right it is somewhat larger. These values are in satisfactory agreement with the theoretical value $\Delta\lambda_2^{(t)}$, calculated in accordance with expression (15) using the formula $\Delta\lambda_2 = \lambda_2/2c\,|\nu|\,l$ ($\Delta\lambda_2^{(t)} = 1.8$ Å for the $LiNbO_3$ crystal at $l = 1$ cm). In the nonstationary regime, the side maxima of the spectral distribution of the harmonic, shown in Fig. 2b, have different values, owing to inexact adjustment of the crystal along the synchronism direction relative to the frequency of the fundamental radiation, which has maximum spectral density. In addition, the minimum value of the intensity in the spectrum of the harmonic is not equal to zero, possibly because of the presence of a definite divergence in the fundamental-radiation beam, leading to certain interactions (5), and in the beam passing through the spectrograph slit.

Figure 4 shows plots of the spectral distribution of harmonic excited in the nonstationary regime by a strongly diverging beam. To this end, a negative lens was placed in front of the $LiNbO_3$ crystal, and a positive lens behind the crystal, to gather the radiation on to the slit of the spectrograph. A comparison of the curves of Fig. 3 with the curves of Fig. 4 shows that in the nonstationary generation regime the spectrum of the harmonic becomes smoother with increasing divergence of the fundamental radiation, and its width increases. Thus, by varying the divergence of the laser beam, it is possible to change the width of the spectrum, meaning also the duration of the harmonic pulse.

The angle structure of the second harmonic is shown in Fig. 5. A periodic structure is clearly seen in the angular distribution of the harmonic generated in the KDP crystal[5)]. No such structure exists in the angular

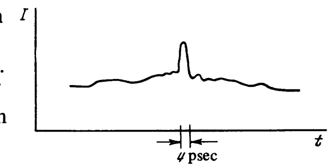


FIG. 1. Intensity of two-photon fluorescence excited by laser radiation ($\lambda_1 = 1.06\ \mu$) in rhodamine 6G. The figure indicates the duration of the ultra-short pulse calculated from the spatial dimension of the bright spot of fluorescence.

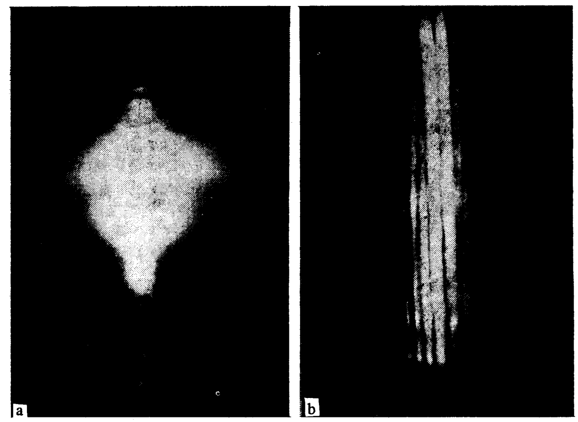


FIG. 2. Spectral distribution of second harmonic, generated by an almost plane-parallel beam of fundamental radiation: a — in quasistatic regime (KDP crystal, $l < l_q$), b — in nonstationary regime ($LiNbO_3$ crystal, $l > l_q$).

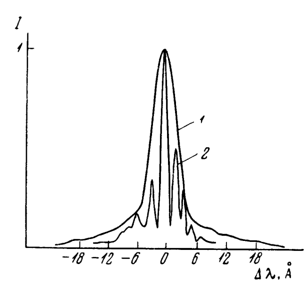


FIG. 3

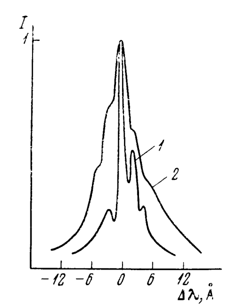


FIG. 4

FIG. 3. Photometry curves: 1 — case of Fig. 2a, 2 — Fig. 2b.

FIG. 4. Experimental plots of the spectral distribution on the second harmonic excited in the nonstationary regime in the $LiNbO_3$ crystal by a diverging beam formed by lenses with focal lengths $R_1 = -25$ cm (curve 1) and $R_1 = -10$ cm (curve 2).

5) Exactly the same angular structure was observed earlier in the harmonic generated by radiation with a narrow spectrum [15].

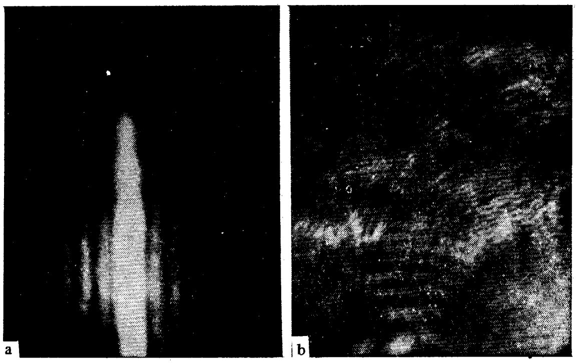

FIG. 5. Angular distribution of second harmonic generated by a diverging fundamental-radiation beam: a — in the quasistatic regime (KDP, $l < l_q$), b — in the nonstationary regime ($LiNbO_3$, $l > l_q$).

distribution of the harmonic emerging from the $LiNbO_3$ crystal, and the distribution here is almost homogeneous[6)]. The results are in agreement with the theory of Sec. 1.

We also investigated experimentally the energy characteristics of a frequency doubler for picosecond pulses. At small coefficients of conversion of the laser radiation into the harmonic, the conversion in $LiNbO_3$ ($l$ = 1 cm) was almost twice as effective as in KDP ($l$ = 2.5 cm); this value is close to the calculated one.

## CONCLUSION

We can thus draw the following conclusions from our experimental and theoretical investigations of second-harmonic derivation by beams of laser radiation of picosecond duration. The frequency spectrum of the harmonic in the nonstationary generation regime, in the absence of effects connected with spatial boundedness and divergence of the laser radiation and with the anisotropy of the nonlinear crystals, has a discrete-continuous spectrum (it is modulated like $\sin^2 x/x^2$. When these effects are significant, the spectral distribution of the harmonic becomes smoothed out continuous. The width of the harmonic spectrum is determined here not only by the quasistatic width, but also by the ratio of the characteristic quasistatic and aperture (or its equivalent) lengths. If the spatial effects become manifest at shorter lengths than the effect of the temporal nonmonochromaticity and of the dispersion of the crystal, then the generation of the harmonic occurs as in the quasistatic regime.

The angular spectrum of the harmonic behaves similarly. If the temporal effects do not influence the process of harmonic generation, then the angular spectrum of the harmonic is discrete-continuous; in the opposite case (in the nonstationary regime) the angular distribution is continuous. In the nonstationary generation regime, the space-time structure of the harmonic beam differs strongly from the structure of the laser radiation.

The dependence of the widths of the frequency and angular spectra of the harmonic in the nonstationary regime on the parameters of the laser radiation can obviously be used to obtain harmonics with specified parameters. Experimentally, it is easiest to realize the dependence of the frequency spectrum of the harmonic, and consequently of the pulse duration, on the angular divergence of the laser radiation, which can be readily varied.

Finally, we note that in the nonstationary generation regime, the maximum energy of the harmonic at optimal focusing depends on the space-time structure of the laser beam.

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6) The rings seen on the photographs of Fig. 5b are due to interference of the light scattered by the inhomogeneity of the crystal; similar rings are observed in the field of the radiation of a single-mode gas laser when this radiation passes through the crystal.

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