WAVE PATTERN OF THIRD OPTICAL HARMONIC GENERATION IN ISOTROPIC AND ANISOTROPIC MEDIA

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Results of a theoretical and experimental investigation of third harmonic generation (THG) in media with an inversion center are reported. THG in isotropic and anisotropic media is considered in the quasi-optical approximation. Special attention is paid to wave effects, namely to the angular spectrum of the harmonic in converging and diverging beams, and to the influence of time modulation of the fundamental radiation on the spatial structure of the harmonic. A specific feature of THG is that an important role may be played by self-actions that result from the same nonlinearity that causes frequency tripling. It is demonstrated that self-action of waves may lead to an appreciable decrease of the tripling efficiency. Optimal conditions for focusing the fundamental radiation during THG in isotropic and anisotropic media are determined. Specific effects involving the $o_1o_1e_1 \rightarrow e_3$ interaction and manifest in the fact that the harmonic angular spectrum is sensitive to variation of the ordinary and extraordinary wave phases of the fundamental radiation, are investigated theoretically and experimentally. A frequency tripler for a single-mode laser with an output power of 1 Mw/cm^2 is described. This is the highest third harmonic power obtained up to present.

1. INTRODUCTION

THE first experimental investigations of third-harmonic generation (THG) in crystals with cubic nonlinearity were performed back in 1963–64[1]; the authors of these investigations confined themselves, however, only to a determination of the nonlinear susceptibilities of third order of different crystals, and to a determination of the synchronism conditions. The theory of THG, which was developed in approximately the same years (see, for example,[2,3]), did not go beyond the frame of geometrical optics of homogeneous beams. In such an approximation (essentially for plane unmodulated waves), an analysis was made of the dependence of the process of frequency tripling on the detuning of the wave vectors, the attenuation of the waves in the medium, the self-action of the waves (nonlinear detuning), and the reaction of the harmonic on the pump.

At the present time, investigations of THG acquire an increasing practical interest in connection with the development of lasers with synchronized modes, which make it possible to obtain such light-field intensities in which cubic effects become comparable with the quadratic effects. Recent investigations have also extended the list of materials in which THG is observed[4-6]. In this connection, undisputed interest attaches to the development of the wave theory of THG, in which account is taken of the dispersion properties of the nonlinear medium, and the inevitably-present spatial and temporal modulation of the fundamental radiation. A detailed analysis of THG, which is presented below, shows that the wave picture of frequency tripling differs significantly from that for the second harmonic.

1. It is very important, in particular, that cubic nonlinearity can produce simultaneously with the frequency tripling also a large number of other nonlinear effects, primarily self-action of the waves, which leads to a change of the phase velocities, and consequently to violation of the synchronism conditions, which in final analysis can lead to a significant limitation of the efficiency of the tripler.

2. The power of the higher harmonics depends quite strongly on the parameters of the laser beam. The energy of the N-th harmonic, $W_N \sim W_1^N L^2(a^2\tau)^{1-N}$ (a—radius of beam, L—length of nonlinear medium, τ—pulse duration). The strong dependence of W_3 on the radius of the beam leads to essentially other conditions of frequency multiplication in anisotropic media than in the case of the second harmonic.

3. In the spatial-angular structure and in the frequency spectrum of the third harmonic (TH) there are also distinct features compared with second harmonics. For example, the fine structure of the angle spectrum of the TH is much less pronounced when a laser beam is focused.

4. We note, finally, that TH can be generated in synchronism not only in anisotropic but also in isotropic media[5].

2. FORMULATION OF PROBLEM. GENERAL EQUATIONS

The analysis of the THG process will be carried out on the basis of the parabolic equations for slowly varying amplitudes[7]. In the given-field approximation, THG in a cubic medium is described by the equations

$$\frac{\partial A_{1o}}{\partial z}+\frac{i}{2k_{1o}}\left(\frac{\partial A_{1o}{}^2}{\partial x^2}+\frac{\partial^2 A_{1o}}{\partial y^2}\right)-\frac{ig_{1o}}{2}\frac{\partial^2 A_{1o}}{\partial\eta^2}+i\sigma_{1o}|A_{1o}|^2A_{1o}+\ldots=0, \quad (1)$$

$$\frac{\partial A_{1e}}{\partial z}+\beta_1\frac{\partial A_{1e}}{\partial x}+\nu_1\frac{\partial A_{1e}}{\partial\eta}+\frac{i}{2k_{1e}}\left(\frac{\partial^2 A_{1e}}{\partial x^2}+\frac{\partial^2 A_{1e}}{\partial y^2}\right)-\frac{ig_{1e}}{2}\frac{\partial^2 A_{1e}}{\partial\eta^2}+i\sigma_{1e}|A_{1e}|^2A_{1e}+\ldots=0, \quad (2)$$

$$\frac{\partial A_{3e}}{\partial z}+\beta_3\frac{\partial A_{3e}}{\partial x}+\nu_3\frac{\partial A_{3e}}{\partial\eta}+\frac{i}{2k_{3e}}\left(\frac{\partial^2 A_{3e}}{\partial x^2}+\frac{\partial^2 A_{3e}}{\partial y^2}\right)-\frac{ig_{3e}}{2}\frac{\partial^2 A_{3e}}{\partial\eta^2}+i\sigma_{3o}|A_{1o}|^2A_{3e}+\ldots=-i\sigma A_{1o}{}^2A_{1e}\exp[-i\Delta_{\text{л}}z], \quad (3)$$

where the z axis is directed along the wave propagation; the indices o and e pertain to the ordinary and extraordinary waves; $\eta = t - z/u_{10}$; $\nu_1 = u_{1e}^{-1} - u_{10}^{-1}$ are the group-velocity detunings; $g = \partial^2 k/\partial\omega^2$ is the group-velocity dispersion coefficient; β is the anisotropy angle between the ray and the wave vectors of the wave; $\Delta_l = k_3 - 2k_{10} - k_{1e}$ is the linear detuning of the wave vector:

$$\sigma = \frac{6\pi\omega}{cn}(e_3\hat{\theta}e_{1o}e_{1o}e_{1e})$$

(similar expressions can be written also for the remaining coefficients σ_{io} and σ_{ie}), e are the polarization vectors, and $\overline{\theta}$ is the cubic nonlinear susceptibility.

We consider THG by a Gaussian beam. The laser operates at the lowest transverse mode TEM_{00q} of a confocal resonator, and this mode experiences linear frequency modulation (FM)[1)]. The beam is focused by a spherical lens of focal length R:

$$A_1(x, y, 0, t) = E_1 \exp\left\{-\left(\frac{1}{a^2} - \frac{ik_1}{2R}\right)(x^2 + y^2) + i\gamma t^2\right\}. \qquad (4)$$

We assume that $A_3 = 0$ on the boundary of the linear medium.

Although it is difficult to obtain a complete analytic solution of the system (1)—(3), an analysis of different particular cases makes it possible to obtain a rather complete picture of the frequency-tripling process.

3. INFLUENCE OF WAVE SELF-ACTION ON THE THG

Let us consider first a bounded light beam, unmodulated in time, and let us assume that the self-action occurs on the same nonlinearity (electronic) as the THG. Then, according to[8,9], the result of the self-action is self-focusing of the beam. During the self-focusing process, two effects are produced and can noticeably influence the THG: 1) the shapes of the wave fronts of the fundamental wave and of the harmonic wave change, 2) narrowing of the beam takes place at the length $L \geq Z_{sf} \approx L_d(P/P_{cr} - 1)^{-1/2}$, where $P_{cr} = cn^2/16\sigma_1 k_1$ and $L_d = k_1 a^2/2$. This causes a noticeable increase of the field intensity of the light wave. We note first that the "total" transfer of the power of the fundamental wave into the power of the harmonic occurs at a length $L_{nl} \approx (\sigma E^2)^{-1}$ [2,3]. A comparison of the length

$$L_{nl}/Z_{sf} \approx (P/P_{cr} - 1)^{1/2} P_{cr}/P$$

shows that we always have $L_{nl} \leq Z_{sf}/2$, and when $P \gg P_{cr}$ we have $L_{nl} \ll Z_{sf}$, i.e., the narrowing of the beam due to self-focusing has practically no effect on the transfer of the fundamental radiation to the TH.

Solving the system (1)—(3) in the geometrical-optics approximation (we disregard the second derivatives with respect to x, y, and η), assuming at the same time that the medium is weakly anisotropic ($\beta \approx 0$), we obtain for the intensity of the harmonic I_3

$$I_3 = \sigma^2 I_1^3 L^2 \frac{\sin^2(\Delta L/2)}{(\Delta L/2)^2}, \qquad (5)$$

$$\Delta = \Delta_l + \Delta_{nl}, \quad \Delta_{nl} = (3\sigma_1 - \sigma_3) I_1.$$

If I_1 is constant over the cross section of the beam (the phase front remains plane), then, choosing $\Delta_l = -\Delta_{nl}$, we can compensate for the nonlinear detuning, obtaining $\Delta = 0$ and $I_3 = \sigma^2 I_1^3 L^2$. In real cases, however, the amplitude is not constant over the cross section of the beam, so that complete compensation of $\Delta_{nl}(r)$ is impossible.

For a Gaussian beam, the third-harmonic power is

$$P_3 = \frac{2\sigma^2 P_1}{(3\sigma_1 - \sigma_3)^2} f(\psi_l, \psi_{nl}), \qquad (6)$$

where the function f takes into account the influence of the detunings

$$f = \frac{\psi - \sin\psi}{\psi_{nl}} + \frac{\psi_l}{\psi_{nl}}\left[\frac{\sin\psi_l}{\psi_l} - 2\ln\left|\frac{\psi}{\psi_l}\right| + 2\,\mathrm{Ci}\,\psi - 2\,\mathrm{Ci}\,\psi_l - \frac{\psi_l}{\psi}(1 - \cos\psi) - \cos\psi_l + \psi_l(\mathrm{Si}\,\psi - \mathrm{Si}\,\psi_l)\right]; \qquad (7)$$

here $\psi = \psi_l + \psi_{nl}$, $\psi_l = \Delta_l L/2$, $\psi_{nl} = \Delta_{nl}(0)L/2$. When $\Delta_l = 0$ and $\psi = \psi_{nl}$ we get from (7) $f = 1 - \sin\psi_{nl}/\psi_{nl}$.

Formula (6) enables us to trace the variation of the efficiency of the tripler with changing power of the fundamental radiation P_1. The critical parameter is here the quantity

$$\psi_{nl} \approx \frac{P_1}{P_0} \approx \frac{L}{L_{nl}} \quad \left(P_0 = \frac{cna^2}{16\sigma L}\right).$$

When $P_1 \gg P_0$, the efficiency of the tripler becomes effected by the change of the phase fronts as a result of the self-action, and this leads to a limitation of the power of the harmonic in the given field of the fundamental radiation, at the level

$$P_3 = \frac{2\sigma^2}{(3\sigma_1 - \sigma_3)^2} P_1. \qquad (8)$$

Numerical estimates yield $P_3 \approx 0.25\, P_1$. We point out for comparison that the theory of plane waves yields in this case $P_3 = 0.6\, P_1$ [2]. The foregoing illustrates the importance of taking into account the beam profile in the analysis of the effects of nonlinear detuning.

The harmonic power can be increased by introducing a "compensating" linear detuning. For example, if we compensate for the nonlinear detuning at the maximum of the amplitude ($\psi = 0$), then the power of the harmonic increases in proportion to the square of the power P_1 and the length L (Fig. 1).

In determining the energy efficiency of the triplet, it is necessary to take into account the time variation of the pump power $P_1(t)$. For example, for a triangular (in intensity) pulse of duration τ and for a Gaussian beam, when $\psi_l = 0$, we have for the energy of the harmonic

$$W_3 = \frac{2\sigma W_1}{(3\sigma_1 - \sigma_3)^2}\left(1 - \frac{\sin\psi_{nl}/2}{\psi_{nl}/2}\right) \qquad (9)$$

For large ψ_{nl}, as for the power P_3, saturation of the harmonic energy takes place. Thus, allowance for the self-action is essential when $\psi_{nl} \gtrsim 1$ or $P \gtrsim \sigma P_0/\sqrt{3}(3\sigma_1 - \sigma_3)$.

It should be noted that the nonlinearities σ_i which are responsible for the self-action can be due also to the Kerr effect, electrostriction, or heating, or can be larger by several orders of magnitude than the electronic σ_{th}. The ratio L_{nl}/Z_{sf} has in this case the form

$$\frac{L_{nl}}{Z_{sf}} = \frac{\sigma_{sf}}{\sigma_{th}}\left(\frac{P}{P_{cr}} - 1\right)^{1/2}\frac{P_{cr}}{P}.$$

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1) Such FM occurs in picosecond pulses generated by lasers with synchronized modes; its calculation is presented below (Sec. 6) in the analysis of nonstationary phenomena in THG.

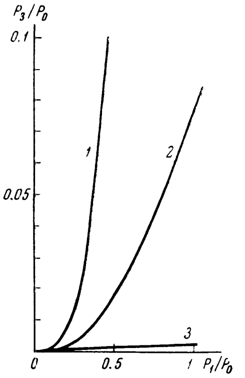

FIG. 1. Dependence of the third-harmonic power P_3 on the pump power P_1: curve 1—in the absence of self-action, $P_3 \sim P_1^3$; 2 and 3—in the case of self-action. Curve 2—the nonlinear detuning is compensated by the linear detuning at the maximum of the amplitude, $P_3 \sim P_1^2$; curve 3—without linear detuning, $P_3 \sim P_1$; P_0—normalization quantity.

If $\sigma_{sf} \gg \sigma_{th}$, a noticeable influence on the THG will be exerted by the change of the power density due to the self-focusing. For example, in liquids with a large Kerr constant, $\sigma_{sf}/\sigma_{th} \approx 10^2$, the self-focusing effects play a decisive role in THG up to $P_1 = 10^4 P_{cr}$.

Further calculations of the harmonic field in isotropic and anisotropic media, using focused beams, pertain to the case of low powers $P_1 \ll P_0$, when the self-action can be disregarded (i.e., the corresponding terms in Eqs. (1)—(3) are omitted).

4. SYNCHRONOUS THG IN ISOTROPIC MEDIA

The condition of wave synchronism in THG in isotropic media can be realized with the aid of specially introduced additives [5], which change the dispersion of the medium in a specified manner. In this case there are no aperture effects at all, making it possible to use quite effectively the accumulating interaction.

The THG process in isotropic media is described by Eqs. (1)—(3), where $\beta_1 = 0$ and $A_{1e} = A_{1o} = A_1$. From (1) and the condition (4) we get

$$A_1 = \frac{E_1}{\xi}\exp\left[-\frac{x^2+y^2}{\xi}\left(\frac{1}{a^2}-\frac{ik_1}{2R}\right)\right], \quad \xi = z\left(\frac{i}{L_d}+\frac{1}{R}\right). \quad (10)$$

The dimensionless width of the beam is determined by the quantity $|\xi|$. At a distance $Z_f = R(\alpha_{10}/\alpha_1)^2$ from the lens, the radius of the beam is minimal, $a_f = 2/k\alpha_1$ (pinching of the beam). Here $\alpha_d = 2/k_1 a$ is the diffraction divergence, $\alpha_{10} = a/R$ is the initial divergence, and $\alpha_1 = (\alpha_{10}^2 + \alpha_d^2)^{1/2}$ is the generalized divergence of the beam in the far field.

Substituting A_1 in (3) and changing over to the spectrum by using the Fourier transformation

$$S(k_{3x}, k_{3y}) = \frac{1}{(2\pi)^2}\iint_{-\infty}^{\infty} A_3(x,y)\exp(ik_{3x}x + ik_{3y}y)\,dx\,dy,$$

we can obtain the distribution of the intensity of the harmonic in the far field:

$$I_3(\theta,\varphi) = \frac{256 n^2\sigma^2 P_1{}^3 \alpha_d{}^2}{9\pi^2 c^3 \alpha_1{}^2 a^2}\exp\left(-\frac{6(\theta^2+\varphi^2)}{\alpha_1{}^2}\right)F, \quad (11)$$

where $\theta = k_{3x}/3k_1$ and $\varphi = k_{3y}/3k_1$ are the observation angles. The quantity

$$F = \left|\int_{L_1}^{L_2}\frac{\exp(-i\Delta_l z)}{\xi^2(z)}\,dz\right|^2 \quad (12)$$

takes into account the violation of the synchronism as a result of the linear detuning Δ_l, and as a result of the additional phase advance of the converging wave (amounting to π in the limit) an effective decrease of the wave number k, by an amount $\Delta k_f = \pi k\alpha_{10}^2/4$, takes place in the focal region, leading to violation of the conditions for one-dimensional synchronism. L_1 and L_2 determine the position of the nonlinear medium, $L = L_2 - L_1$.

The degree of focusing of the beam will be characterized by the focusing parameter

$$m = L/L_f, \quad (13)$$

where $L_f = 4/k_1\alpha_1^2$ is the length of the focal spot. The angular distribution of the intensity of the harmonic, as seen from (11), remains Gaussian but subtends a solid angle smaller by a factor of 3 than the fundamental radiation.

The power of the harmonic in the case of exact synchronism $\Delta_l = 0$ is

$$P_3 = \frac{4P_{30}m^2}{[1+4m^2(1-q)^2][1+4m^2q^2]}. \quad (14)$$

Here $P_{30} = 16\sigma^2 n^2 P_1^3 k^2/3\pi^2 c^2$, $q = l/L$ is the relative distance of the pinch from the front boundary of the nonlinear medium, $l = Z_f - L_1$, at $l = 0$ the beam is focused on the front boundary and at $l = L$ on the rear boundary. The power P_3 for a beam with diffraction divergence corresponds to $R \to \infty$; for $L_1 = 0$ we have $P_3 = P_{30}L^2/(L^2 + L_d^2)$. In TH generation by a parallel beam, $L \ll L_d$ and $P_3 = P_{30}L^2/L_d^2$.

For a given lens with a focusing parameter m we can indicate the optimal location of the nonlinear medium, at which the harmonic power is maximal. In the case $m \le 1$ we get from (14) $q = 1/2$ (the pinching is in the middle of the medium);

$$P_3 = P_{30}\frac{4m^2}{(1+m^2)^2} \le P_{30}.$$

When $m \ge 1$ we have for the position of the focus $q = 1/2(1 \pm \sqrt{1-m^2})$, when $P_3 = P_{30}$.

As already indicated, the synchronism condition is violated at the focus, and to increase the efficiency it is desirable to introduce a linear detuning $\Delta_l > 0$. For example, in the case of strong focusing of the beam ($m \gg 1$) in the center of the medium ($P_3 = P_{30}L_d^{-2}F$) it follows from the asymptotic expression for the function F(12)

$$F \approx L_d{}^2 m^{-2}[2\cos\psi_l + 2\psi_l \operatorname{Si}\psi_l + (1+\operatorname{sign}\Delta_l)\pi\psi_l\exp(-\psi_l/m)]^2 \quad (15)$$

that now the maximum of the power is reached at $\Delta_l = 2/L_f$ (see Fig. 2). Compared with $\Delta_l = 0$, a gain is

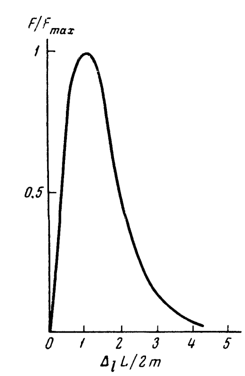

FIG. 2. The function F, which characterizes the THG in focused beams. It gives also the angular distribution of the harmonic for the interaction $o_1 o_1 o_1 - e_3$.

short-focus lenses are used it is more convenient to generate the harmonic with additional phase detuning.

5. THG IN ANISOTROPIC MEDIA

Synchronous interactions in THG can be realized also in transparent anisotropic media between waves of different polarization. Unlike isotropic media, in anisotropic media there are strictly defined synchronism directions. The difference between the refractive indices of the crystal in the other directions leads to a strong influence of the divergence and limited character of the beam on the THG efficiency (see papers on second-harmonic generation, for example,[10,11]). We analyze below the character of the variation of the quantities I_3 and P_3 as functions of the ratios of the parameters of the beam, of the lens, and of the crystal for different types of interaction.

1. Interaction $o_1o_1o_1 - e_3$(I)

For the interaction (I) it is necessary to put in (1)–(3) $\beta_1 = 0$ and $A_{1e} = A_{1o}$. Integrating (1)–(3) for the intensity of the angle spectrum, we obtain the previous formula (11), except that Δ_l is replaced by $\Delta_l + 3k\beta_3\theta$, which takes into account the anisotropy of the medium.

a) Generation by a weakly-divergent beam ($L \ll L_d$, R). In this case the amplitude profile of the fundamental radiation remains practically unchanged over the length of the crystal as a result of the divergence, and in the integral (12) we can set the denominator equal to a constant. Then, at $\Delta_l = 0$ we have

$$F=\frac{L^2}{[(1-L_1/R)^2+L_1^2/L_d^2]^2}\frac{\sin^2\bar{\theta}}{\bar{\theta}^2}, \qquad (16)$$

where $\bar{\theta} = \theta/\theta_c$, $\theta_c = 2(3k_1\beta_3L)^{-1}$. It is obvious that bands as functions of the angle θ will appear in the angle structure of the harmonic, if $\alpha_1 > \theta_c$. This can be due either to the angular aperture effect, when the initial divergence of the beam $\alpha_{10} > \alpha_c$, or as a result of the diaphragm aperture effects, when the aperture length $L < L$, $L_\beta = (3/2)^{1/2}a/\beta_3$. In the near field, at the exit from the nonlinear crystal, in the case of the angle aperture effect, the field of the harmonic also has the form of bands, and in the case of the diaphragm aperture effect the beam of the harmonic in the x direction is broader than the fundamental beam by a factor L/L_β.

The harmonic power depends on the length of the crystal in accordance with the law

$$P_3=\frac{P_{30}L^2\zeta^{-1}}{L_d^2[(1-L_1/R)^2+L_1^2/L_d^2]^2}\left[\sqrt{\pi}\,\Phi(\zeta)+\frac{\exp(-\zeta^2)-1}{\zeta}\right], \qquad (17)$$

where $\Phi(\zeta)$ is the error function, $\zeta = L/l_\beta$, and $l_\beta = L_\beta\alpha_d/\alpha_1$ is the generalized aperture length, which takes into account both the drift and the divergence of the beam. We see that when $\xi \ll 1$ the power is $P_3 \sim L^2$, just as in an isotropic medium. When $\xi \gg 1$, the aperture effects decrease the power, $P_3 \sim L l_\beta$. With increasing distance from the crystal to the laser (as L_1 increases), the power of the harmonic decreases.

b) Focusing of beam by a spherical lens. Let us consider THG when the laser beam is focused in the middle of a crystal ($L_1 = R - L/2n$) by a spherical lens with focal length $R \ll L_d$.

The intensity of the harmonic in the far field is given by

$$I_3(\theta,\varphi)=\frac{64\sigma^2P_1^3nk_1L}{9\pi^2c^3m^3}\exp\left(-\frac{6(\theta^2+\varphi^2)}{\alpha_1^2}\right)F_1,$$

$$F_1=\left|\int_{-1}^{1}\frac{\exp(-i\tau\bar{\theta})}{(\tau+i/m)^2}d\tau\right|^2. \qquad (18)$$

In the case of focusing of a laser beam by a long-fosused lens ($m \ll 1$) we obtain from (18) formulas similar to (16) and (17), describing harmonic generation by a parallel beam of radius $a_f = 2/k_1\alpha_0$. If the focusing angle is $\alpha_0 > \theta_c$ (or if $L > a_f/\beta_3$), then the angle pattern of the harmonic has the form of symmetrical bands in terms of the angle θ with an angle width $\pi\theta$.

When the beam is focused by a short-focus lens ($m \gg 1$), the aperture function has an asymptotic expression obtained from (15) by making the substitutions $\psi_l \to \theta$ and $\Delta_l \to \theta$. The fine structure of the angle spectrum is weakly pronounced (see Fig. 2), and the radiation is contained in practice in the region of the vector synchronism $\theta > 0$[2].

In the case when the influence of the aperture effect can be neglected, i.e., $\min(L, L_f) \ll a_f/\beta_3$ or $m \ll m \ll m^{-1}$, where $m_\beta = nk_1L\beta_3^2/4$, the power of the harmonic is determined, just as for an isotropic medium, by expression (14) with $q = 1/2$ (see Fig. 3, curve 1).

If the aperture effects are appreciable, i.e., $\min(L, L_f) \gg a_f/\beta_3$ ($m_\beta^{-1} \ll m \ll m_\beta$), then the power of the harmonic depends on the focusing parameter m in the following manner (Fig. 3, curve 2):

$$P_3=\frac{\sqrt{2\pi}\,P_{30}m^{3/2}}{\sqrt{3}\,m_\beta^{1/2}}\left(\frac{1}{\sqrt{1+m^2}}+\frac{\operatorname{arctg} m}{m}\right). \qquad (19)$$

When $m \gg 1$ (short-focus lens), the power increases in proportion to $\sqrt{m}$ until $m \ll m_\beta$. When $m \gg m_\beta$, it is necessary to use formula (14), from which it follows that when $m \to \infty$ the power decreases like m^{-2}.

Thus, there should be a maximum in the parameter region $m \approx m_\beta$. However, the case $m \approx m_\beta$ entails the greatest difficulties in the integration of the general expression for the power. If $m \gg 1$ and $m_\beta \gg 1$, the aperture function can be written approximately in the form $F_1 \approx 0$ for $\theta < 0$ and in the region $\theta > 0$ we have

$$F_1=\pi^2nk_1Lm^{-3}\bar{\theta}\exp(-2\bar{\theta}/m). \qquad (20)$$

Integration of (18) with allowance for (20) leads to the formula

$$P_3=2\pi\sqrt{\pi}P_{30}\left\{-b^3+\frac{\sqrt{\pi}}{2}b^2(1+2b^2)[1-\Phi(b)]e^{b^2}\right\}, \qquad (21)$$

where $b = (6nm_\beta/m)^{1/2}$. The function has a maximum $P_3 = 1.65P_{30}$ at $b \approx 2$, i.e., at $\alpha_0 \approx n\beta_3$ (Fig. 3, curve 3). Thus, the focal distance of the optimal lens $R = a/\alpha_0 \approx L_\beta$, i.e., it is equal to the aperture length. This condition is conserved for crystals with parameter m_β up to unity. We note also that, unlike SHG, in THG in anisotropic media no loss of power occurs compared

[2]The TH spectrum differs strongly in this respect from the second-harmonic spectrum, which reveals a noticeable fine structure. The reason for this difference is the stronger degree of the dependence of the THG on the light field of the fundamental radiation ($A_N \sim A_1{}^N$), in connection with which the effective phase detunings in the focal region become more significant, since $\Delta_f \approx N\pi$.

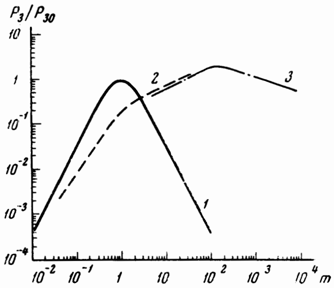

FIG. 3. Dependence of the harmonic power P_3 on the parameter of focusing of the laser beam by a spherical lens in the center of a crystal; interaction $o_1o_1o_1-e_3$. 1—weakly-anisotropic medium, $m_\beta \ll 1$; 2—strongly anisotropic medium, $m_\beta = 100$; $m \ll m_\beta$; 3—the same, but $m \gg 1$.

with the THG in an isotropic medium (in both cases $P_{3,opt} \approx P_{30}$).

c) Focusing of beam with cylindrical lens. Let us consider THG with the beam focused into the center of the crystal by a cylindrical lens at two lens orientations: focusing with respect to the angle θ or with respect to φ. It should be noted that the TH aperture function for a cylindrical lens is analogous to the aperture function of the second harmonic for a spherical lens [11]. We therefore proceed directly to analyze the power of the harmonic.

Let the focusing be with respect to the angle φ (in this case the anisotropic properties of the media are less pronounced and we can obtain a larger gain in power than in focusing with respect to θ). If min(L, L_f) $\ll L_\beta$, i.e., $m_\beta m_d \ll 1$ at $m \lesssim 1$ or $m_d m_\beta \ll m^2$ at $m \gg 1$ ($m_d = L/2L_d$), then the third-harmonic power is

$$P_3 = 4P_{30}m_d m^{-1} \operatorname{arctg} m. \tag{22a}$$

In the case of optimal focusing (m = 1.4) the power reaches a maximum $P_3 = 2.6\, m_d P_{30}$. On the other hand, if min(L, L_f) $\gg L_\beta$, then the power

$$P_3 = \sqrt{\frac{2\pi m_d}{3m_\beta}}\, P_{30} \operatorname{arc\,tg} m$$

increases with increasing degree of focusing until m reaches the value $m \approx \sqrt{m_d m_\beta} = L/L_\beta$. With further increase, the power decreases, in accordance with (22a). Thus, when $L \gg L_\beta$ the maximum power is reached by focusing with a cylindrical lens with $m = \sqrt{m_d m_\beta}$:

$$P_3 = \sqrt{\frac{2\pi m_d}{3m_\beta}}\, P_{30}. \tag{22b}$$

If the beam is focused with respect to the angle θ, if min(L, L_f) $\ll a_f/\beta_3$ ($m_\beta \ll m \ll m^{-1}$), the power and its maximum are expressed by formulas (22). The optimal focusing m = 1.4 can in this case be observed in crystals with a parameter $m_\beta \ll 1$. For crystals with $m_\beta \gg 1$, an important role in optimal focusing of the beam is played by the aperture effects. The power

$$P_3 = \frac{2\sqrt{2\pi}\, P_{30}}{\sqrt{3m_\beta m}} \operatorname{arc\,tg} m$$

has a maximum at m = 1.4: $P_3 = 2.2 m_d m_\beta^{1/2} P_{30}$, which is worse by a factor $m_\beta^{1/2}$ than the maximum obtained by focusing in the angle φ, if $L \ll L_\beta$.

2. Interaction $o_1o_1e_1 - e_3$ (II)

Interaction (II) differs from interaction (I) in that now there propagate in the crystal two beams of fundamental frequency, with ordinary and extraordinary polarization. These beams diverge in space as a result of the birefringence effect, and generation of the harmonic occurs in a limited volume, and not in the entire length of the crystal as in interaction (I).

a) Field of harmonic generated by weakly diverging beams. Such a field can be calculated in the geometrical-optics approximation without taking into account the second derivatives in Eqs. (1)—(3); we then obtain

$$I_3(\theta,\varphi) = \frac{256\sigma^2 P_{1o}^2 P_{1e}\alpha_{1d}\alpha_{2d}}{9\pi^2\alpha_1\alpha_2 c^3 n^3 a_1 a_2} \exp\left(-\frac{6\theta^2}{\alpha_1^2} - \frac{6\varphi^2}{\alpha_2^2}\right) F_2. \tag{23}$$

The aperture function F_2 is expressed in terms of the error function of complex argument (we assume that $\beta_1 = \beta_3$)

$$F_2 = \frac{\pi\alpha_{1d}L_\beta^2}{4\alpha_1} \exp\left(-\frac{12\theta^2}{\alpha_1^2}\right) |\Phi(\eta_1+\eta_2) - \Phi(\eta_2)|^2, \tag{24}$$

where

$$\eta_1 = \frac{L}{L_\beta}\left(1 + i\frac{\alpha_{10}}{\alpha_{1d}}\right)^{1/2}, \quad \eta_2 = i\sqrt{6}\frac{\theta}{\alpha_{1d}}\left(1 + i\frac{\alpha_{10}}{\alpha_{1d}}\right)^{-1/2}.$$

If the length of the anisotropic crystal is such that the ordinary and extraordinary beams do not split from each other, $L \ll L_\beta$, then formula (24) can be represented in the form

$$F_2 = \frac{\pi\alpha_{1d}L_\beta^2}{2\alpha_{10}} [[C(\alpha_e+\theta_e) - C(\theta_e)]^2 + [S(\alpha_e+\theta_e) - S(\theta_e)]^2]. \tag{25}$$

Here C and S are Fresnel integrals;

$$\theta_e = \frac{\sqrt{6}\,\theta}{\sqrt{\alpha_{10}\alpha_{1d}}}, \quad \alpha_e = \sqrt{\frac{\alpha_{10}}{\alpha_d}}\frac{L}{L_\beta}.$$

A plot of this function is shown in Fig. 4, curve 1. This curve lies mainly in the region $\theta < 0$ (closer to the optical axis); it is symmetrical with respect to the direction $\theta = -\alpha_{10}\alpha_d/18\theta_c$. We see that with increasing beam divergence α_{10} the curve shifts more and more and broadens ($\Delta\theta \approx \alpha_{10}\alpha_d/9\theta_c$).

The asymmetry in the distribution of I_3 relative to the synchronism direction ($\theta = 0$) is explained by the contribution of the two-dimensional or vector interactions upon intersection of the elementary rays belonging to beams with ordinary and extraordinary polarization[3)]. This is confirmed also experimentally (see Sec. 7). On the other hand, if the beams separate as a result of birefringence, $L \gg L_\beta$, then in the case of large beam divergence, $\alpha_{10} \gg \alpha_d$, it follows from (23) and (24) that $I_3 \approx 0$ for $\theta > 0$, and in the region $\theta < 0$ we have

$$I_3 \sim \exp(-18\theta^2/\alpha_1^2). \tag{26}$$

In this case the angle width of the TH radiation relative to the angle θ is smaller by a factor of 6 than the fundamental radiation. With respect to the angle φ, as before, the width is decreased by a factor $\sqrt{3}$. Thus, the TH radiation has the form of an oval spot with a diameter ratio ~ 3.5.

As a result of the separation of the ordinary and

[3)] From the vector-synchronism condition $2\mathbf{k}_{10} + \mathbf{k}_{1e} = \mathbf{k}_{3e}$ for small angles we can find that $\theta_3 = \theta_{10} \pm \sqrt{-2\theta_{10}\beta/3}$ and $\theta_{1e} = \theta_{10} \pm \sqrt{-6\theta_{10}\beta}$. We see therefore that the contribution to the two-dimensional interaction is made by rays with $\theta_{10} < 0$, and with this $\theta_3 < 0$.

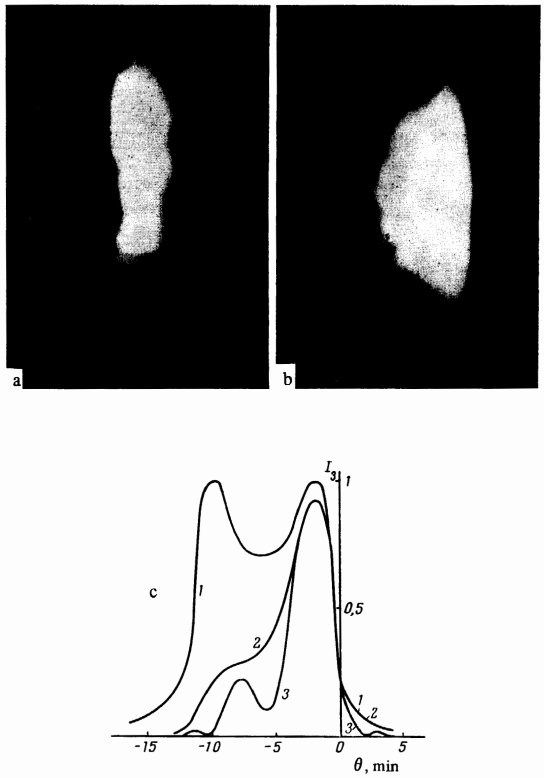

FIG. 4. Distribution of the intensity of the harmonic in the far field. a—THG in diverging beam with $\alpha_1 \approx 30'$; b—THG in focused beam; c—angular spectrum with respect to angle θ (in minutes): 1—aperture function F_2 for $L \ll L_\beta$ (formula (25)); 2—theoretical curve for the intensity I_3 at $L \approx L_\beta$; 3—experimental curve obtained by photometry of photograph a.

extraordinary pump beams ($L \gg L_\beta$), saturation of the harmonic power takes place (even in the given-pump-field approximation):

$$P_3 = \frac{16\sigma^2 P_{1o}^2 P_{1e}\alpha_d}{\pi^2\sqrt{3}\,n^2c^2a^2\beta^2\alpha_1}\left(\pi - \operatorname{arctg}\frac{\sqrt{3}\,\alpha_d}{\alpha_1}\right). \qquad (27)$$

The saturation power, as seen from (27) is quite sensitive to the divergence of the laser beam, namely, it decreases rapidly with increasing initial divergence if the latter exceeds the diffraction value, $P_3 \sim \alpha_{10}^{-1}$ (see Fig. 5). This circumstance, which is inherent in the $o_1e_1e_1 - e_3$ interaction, is very significant (for comparison we recall that in the $o_1o_1o_1 - e_3$ interaction the divergence has an influence if it is larger than θ_c (see (16)).

b) Focused beams. If the length of the focal spot is larger than the length of the crystal, then everything

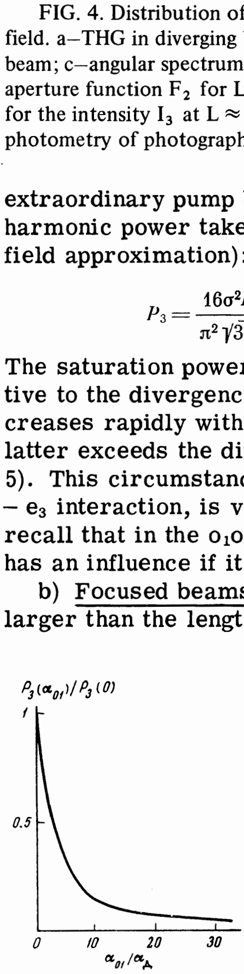

FIG. 5. Dependence of the saturation power of the harmonic ($L \gg L$) on the ratio of the initial beam divergence α_{10} to the diffraction divergence α_d in the interaction $o_1o_1e_1 - e_3$.

proceeds in the same manner as in the generation of TH by parallel beams. The harmonic power, starting with $a_f \leq \beta L$, in accordance with (27), decreases when $\alpha_{10} = 0$, $\alpha_1 = \alpha_d$, and $a = a_f$ in inverse proportion to the cross section of the focal spot. Such an increase of P_3 continues up to focusing with $m \approx 1$. The subsequent process depends on the location of the focus in the crystal.

If the beam is focused in the middle of the crystal, then already at $m \gtrsim 1$ the focal spots corresponding to the ordinary and extraordinary waves practically do not overlap, and the power P_3 decreases sharply with increasing m. Thus, when the beam is focused in the interior of the crystal, the maximum power corresponds to focusing $m \approx 1$:

$$P_3 = \frac{32\sigma^2 P_{1o}^2 P_{1e} k}{3\sqrt{3}\,\pi n^2 c^2 \beta^2 L}. \qquad (28)$$

The gain in power compared with the parallel beam amounts to ka^2/L times at $L > L_\beta$ and ka^4/β^2L^3 times at $L < L_\beta$.

When the beam is focused on the front face of the crystal, the decrease of the radius of the focal spot a_f is advantageous so long as $a_f \approx \beta L_f$, i.e., $m \approx m_\beta$ or $\alpha \approx \beta$. This condition is analogous to the focusing condition in the interaction $o_1o_1o_1 - e_1$ (see (21) and Fig. 3). The gain in power relative to the unfocused beam amounts to $(L_d/L)^2$ times when $L < L_\beta$ and $(\beta/\alpha_d)^2$ times when $L > L_\beta$.

6. NONSTATIONARY PHENOMENA IN THG

Modulation of the laser radiation in time causes the THG process to become nonstationary. For nonresonant nonlinear processes, such as harmonic generation, an important role is assumed first of all by the group delay of the wave packets, and then also by their dispersion spreading.

According to the general equations (1)—(3), the detuning of the group velocities ν is significant at lengths exceeding the quasistatic length $L_\nu = \tau/|\nu|$, and the spreading of the packet occurs at lengths exceeding $L_{sp} = \tau^2/2g$; here τ is the characteristic modulation time. For a calcite crystal the dispersion is such that when $\lambda_1 = 1.06\ \mu$ we have $\nu_{13} = 1.3 \times 10^{-12}$ sec/cm and $g = 10^{-27}$ sec^2/cm, so that at a pump pulse duration $\tau = 10^{-12}$ sec the characteristic lengths are $L_\nu = 0.8$ cm and $L_{sp} = 100$ cm.

If the crystal length is less than the quasistatic length $L < L_\nu$, then we can use in the calculation of the THG the quasistatic approximation, in which the time derivatives in (1)—(3) are disregarded and the time enters in the formulas as a parameter. In this case, for example, we see immediately that the harmonic pulse will be shorter by a factor $\sqrt{3}$ than the laser pulse.

At larger lengths, when $L > L_\nu$, the nonstationary THG theory should be constructed on the basis of the general solutions of the system (1)—(3).

The nonstationary theory is easiest to develop for THG in isotropic media ($\beta = 0$) by using the results already obtained by us. In this case, comparing the nonstationary equations with the equations describing the THG by spatially-modulated beams in anisotropic media, we can develop the space-time analogy[12].[4)] Thus, using the already available formulas, we can immediately obtain from them expressions for the harmonic fields generated by short pulses. However, such an analogy can be drawn only so long as the spatial and temporal modulations of the fundamental wave are each separately of importance to the THG.

In the general case, for waves modulated simultaneously both in space and in time (narrow beams, short pulses), the angular and frequency spectra of the harmonic turn out to be interrelated[5)]. Calculations of the characteristics of the harmonic field are similar to those made above, but now it is necessary to solve the complete system (1)—(3). We present, for example, the results of calculations performed in the quasioptic approximation. The frequency-angle spectrum of the harmonic is given by

$$I_3(\theta, \varphi, \Omega) \sim \exp\left(-\frac{6(\theta^2+\varphi^2)}{\alpha_1{}^2} - \frac{6\Omega^2}{\Omega_1{}^2}\right)\frac{\sin^2(\tilde{\theta}-\tilde{\Omega})}{(\tilde{\theta}-\tilde{\Omega})^2}, \quad (29)$$

where

$$\tilde{\Omega} = \Omega/\Omega_c, \ \Omega_c = 2/\nu L.$$

Gathering the entire TH radiation in the focus of a long-focus length, we can investigate the harmonic pulse (with respect to total power). The corresponding frequency spectrum, obtained from (29) by integrating with respect to all θ and φ, can be written in the following manner:

$$P_3(\Omega) \sim \exp\left(-\frac{6\Omega^2}{\Omega_1{}^2}\right)\left\{\frac{\sqrt{\pi}}{2}\exp\left(-\frac{27\tilde{\Omega}^2\theta_c{}^2}{4\alpha_1{}^2}\right)\left[\zeta\Phi(\zeta)+\zeta^*\Phi(\zeta^*)\right.\right.$$
$$\left.\left. + \frac{2\sqrt{6}\,\tilde{\Omega}\theta_c}{\alpha_1}\operatorname{Erfi}\left(\frac{2\sqrt{6}\,\tilde{\Omega}\theta_c}{\alpha_1}\right)\right] + \exp\left(-\frac{4\alpha_1{}^2}{27\theta_c{}^2}\right)\cos(2\tilde{\Omega}) - 1\right\} \quad (30)$$

where $\zeta = L/l_\beta + i\Omega\tau l_\beta/l_\nu$; l_ν is the quasistatic length for a Gaussian frequency-modulated pulse; $l_\nu = L_\nu(1+\gamma^2\tau^4)^{-1/2}$.

An analysis of expression (30) shows that if $L > l_\nu$, l_β but the predominant role is played by the aperture effects connected with the spatial modulation of the beam, i.e., $l_\beta < l_\nu$, then the spectrum of the TH does not become narrow, as was in the case of generation of TH of a plane wave (see (29)). In the opposite case $l_\nu < l_\beta$, the harmonic spectrum is given by $\Omega_3 \sim 2\pi/\nu L$. Thus, by varying the spatial modulation of the beam (its radius or its divergence) it is possible to vary the temporal modulation of the harmonic (the pulse duration).

The energy of the harmonic changes as a function of the crystal length in the following manner (compare with (17)):

$$W_3 = \frac{512\sigma^2 W_1{}^3 L^2\pi^5}{\sqrt{3}\,c^2 n_1{}^2 a^4 \zeta\tau^2}\left[\sqrt{\pi}\,\Phi(\zeta) + \frac{\exp(-\zeta^2)-1}{\zeta}\right], \quad (31)$$

where $\zeta = L/l_{\beta\nu}$, $l_{\beta\nu} = (l_\beta^{-2} + l_\nu^{-2})^{-1/2}$ is the generalized aperture length, which takes into account the influence of the spatial and temporal modulation on the THG. The mechanism that limits the rate of growth of energy W_3 with increasing length L is determined by the ratio of the aperture and quasistatic lengths.

7. EXPERIMENTAL INVESTIGATION OF THG

In our experiments we used a Q-switched neodymium laser, working in the regime of the fundamental transverse mode. The distribution of the intensity over the beam cross section was close to Gaussian with a radius a = 1.5 mm, and the divergence of the beam had practically the diffraction value ~ 2′, while the radiation was plane-polarized. The pulse duration at the half-width was τ = 15 nsec.

As the nonlinear element we used a calcite crystal of length L = 3.4 cm. The dispersion characteristics of the calcite admitted of all three types of synchronous interactions: (I)—$o_1o_1o_1 - e_3$, synchronism direction $\theta = 29°$; (II)—$o_1o_1e_1 - e_3$, $\theta = 35°$; (III)—$o_1e_1e_1 - e_3$, $\theta = 50°$. The samples were cut in such a way that the polarization vector of the ordinary wave was directed along the x_0 axis; the x_0z_0 plane was the symmetry plane with respect to the reflection transformation, and z_0 the optical axis.

The angle structure of the harmonic radiation from the crystal was investigated with the aid of a long-focus objective with focal length R = 1600 mm. A detailed investigation of the TH was carried for the interaction (II), which has a relatively large nonlinear coupling coefficient σ[1].

In an unfocused beam, at a pump power density P_1 = 200 MW/cm², a harmonic power $P_3 \approx$ 1 MW/cm² was attained, corresponding to a conversion efficiency of 0.5%.

We investigated further the angle aperture effect (see Sec. 2, Item 2a), and to register this effect more clearly the fundamental beam was focused beforehand to a divergence of 30′. A photograph of the angle spectrum of the harmonic generated in the diverging beam, and the results of its photometry, are shown in Fig. 4. The angular distribution curves of the intensity are given in relative units. Since in the experiment the crystal length was $L \approx L_\beta$, i.e., the beams in the crystal diverge noticeably relative to each other, the picture is close to the case $L \gtrsim L_\beta$ (see formula (26)). Indeed, the angle width of the harmonic, which is equal to ~ 5′, is smaller by a factor of 6 than the divergence of the laser beam (α_1 = 30′). It must also be emphasized that the picture is shifted relative to the direction of synchronism towards the optical axis of the crystal.

We have also performed experiments aimed at finding the optimal focusing conditions. Owing to the breakdown of the crystal, the experiments were performed at pump power levels on the order of $P_1 \approx$ 20 MW/cm², and the beam was focused in the interior of the crystal.

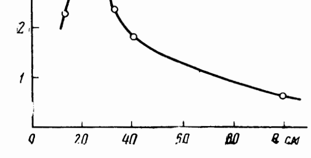

FIG. 6. Experimental dependence of the logarithm of the harmonic power P_3 on the focal distance R of the lens used to focus the beam into the center of the crystal; interaction $o_1o_1e_1 - e_3$.

4)In this problem the main analogs are the following quantities: The anisotropy angle β and the group detuning ν; the beam radius β and the pulse duration τ; the diffraction length L_d and the pulse spreading length L_{sp}; angle spectrum $I_3(\theta, \varphi)$ and the frequency spectrum $I_3(\omega_0 + \Omega)$, etc.

5)This connection was analyzed in detail theoretically and experimentally in the case of second-harmonic generation [13].

The results of the experiment are shown in Fig. 6. As seen from the diagram, the maximum of the power is reached when the beam is focused with a lens of focal length R = 20 cm, which when recalculated in terms of the focusing parameter (13) yields $m \approx 0.8$. The harmonic power gain compared with the unfocused beam is 3×10^3 times. This case corresponds to that considered by us earlier theoretically (Sec. 6, Item 2b). As shown there, optimal focusing corresponds to m = 1, and the power gain does not exceed $(L_d/L)^2$ times, which amounts to $\sim 10^4$ under the conditions of our experiment. Obviously the data obtained experimentally and those calculated theoretically are in good agreement.

We note, finally, that the duration of the harmonic pulse was 9 nsec, i.e., it was smaller by a factor 1.7 compared with the fundamental one. This is also in good agreement with the conclusions of the theory.

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