NONSTATIONARY PHENOMENA AND SPACE-TIME ANALOGY IN NONLINEAR OPTICS

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Nonstationary phenomena in the interaction of short light pulses in media with a polarization quadratic or cubic with respect to the field strength are investigated theoretically. Problems of frequency doubling of picosecond pulses are considered. Group retardation as well as dispersion smearing out of the pulses are taken into account. A similar analysis is performed for parametric amplification in the field of pulsed pumping. The features of cross-modulation of short light pulses in a cubic medium are discussed. Such effects can lead to anomalous broadening of the spectrum. The space-time analogy in the theory of nonlinear interactions of modulated waves is studied in detail. A comparison of equations describing interaction of plane wave packets in a dispersive medium on the one hand, and restricted wave beams non-modulated with respect to time on the other, shows that an analogy can be set up between them by comparing the time derivatives in the former with the derivatives in a plane perpendicular to the ray in the latter. A comparison of the coefficients of the dc ꞉ ˙ ᴗs also has a clear physical meaning. By exploiting the analogy one can apply the results obtained by considering nonlinear interactions in restricted beams to the theory of nonstationary nonlinear effects. Some practical applications of nonstationary effects such as monochromatization of the frequency spectrum, formation of short powerful pulses, etc. are discussed. It is noted in particular that nonstationary phenomena in generation of optical harmonics can be used to investigate the amplitude and phase characteristics of broadband optical signals obtained by self-focusing.

1. INTRODUCTION (NONSTATIONARY EFFECTS IN NONLINEAR OPTICS OF PICOSECOND PULSES; FUNDAMENTAL EQUATIONS)

1. It is known that the results of the theory of nonlinear interactions of plane waves cannot always be used to interpret the experimental data; in real situations, the modulation of the interacting waves plays an important role. In particular, in nonlinear optics, many important features of the interaction of light waves are connected with the spatial modulation (finite aperture of the beams). As to the effects of the time modulation which inevitably is present in laser radiation, they could certainly be neglected up to the very latest time, even in experiments with pulses of duration $\tau_p \approx 10^{-8}$ sec, and the problem could be regarded as stationary[1)]. The situation changed radically after picosecond-pulse generators were developed ($\tau_p \approx 10^{-12}$ sec); here, as shown by estimates, practically all typical nonlinear wave interactions become essentially nonstationary. The nonstationary character of the nonlinear effect can be connected, generally speaking, with two circumstances—the non-quasistatic character of the local nonlinear response (if the relaxation time of the nonlinearity is $\tau > \tau_p$) and the non-quasistatic character of the response of the nonlinear medium as a whole (if the nonlinear effect at the given point of space and given instant of time depends on the values of the initial fields at separated instants of time, then in this case the non-quasistatic character is connected with the group delay of the pulses).

In the present article we consider only nonstationary features of the second type; we are dealing with nonlinear interactions in a low-inertia ($\tau \approx 10^{-15}$ sec) electronic nonlinearity. We describe below a procedure and the results of the solution of several problems of nonlinear optics of ultrashort pulses. The analysis is based on an approximate parabolic equation (the possibility of its utilization for the description of nonstationary processes is discussed in[1,2]). An interesting circumstance revealed in the consideration of the indicated processes is the existence of a sufficiently lucid space-time analogy in the theory of interactions of modulated waves. A comparison of the approximate equations describing the interaction of plane wave packets in dispersive media, on the one hand, and bounded wave beams that are not modulated in time, on the other, shows that a direct analogy can be established between them if the time derivatives in the former are set in correspondence with derivatives in a plane perpendicular to the beam in the latter. In this case, the comparison of the coefficients of these derivatives has also a clear cut meaning[2)]. It thus becomes possible to use many results of the theory of interaction of bounded beams in the theory of nonstationary nonlinear effects.

[1)]It is interesting that this pertains not only to such "rapid" effects as parametric amplification, stimulated Raman scattering (SRS) and harmonic generation, but also stimulated Mandel'shtam-Brillouin scattering (SMBS) in liquids and crystals at room temperature.

[2)]The analogy under consideration has, of course, nothing in common with the well known space-time analogy that establishes the correspondence between the nonlinear interaction of nonmodulated waves and the nonlinear interactions of oscillations in systems with lumped constants (see [3]).

The foregoing does not pertain, of course, to nonlinear optics alone; the considered analogy can be used also in the theory of waves in a plasma, etc. We shall trace below the space-time analogy using as an example processes of two types: nonlinear interactions in a medium with polarization of the type $\mathbf{P} = \hat{\kappa}\mathbf{E} + \hat{\chi}\mathbf{EE}$ (these interactions, for which the phase relations between the waves are very important, we shall call for brevity "carrier interactions"), and interactions in a medium with polarization of the type $\mathbf{P} = \hat{\kappa}\mathbf{E} + \hat{\theta}\mathbf{EEE}$ (principal attention is paid here to interactions for which the phase relations are insignificant; they are called "envelope interactions"). Besides the methodological questions, we consider in the article also new problems, namely the nonstationary generation of the second harmonic under high conversion-efficiency conditions, nonstationary parametric amplification, and cross modulation of light pulses in a nonlinear medium.

It is shown that the nonstationary wave interactions on the electronic nonlinearity can be of appreciable interest towards the shaping of ultrashort pulses and for the study of the amplitude-phase structure of broadband optical signals.

2. The propagation of electromagnetic waves in nonlinear media is described by the equation

$$c^2[\nabla[\nabla\mathbf{E}]] + \frac{\partial^2\mathbf{E}}{\partial t^2} + 4\pi\frac{\partial^2\mathbf{P}^{1}}{\partial t^2} + 4\pi\frac{\partial^2\mathbf{P}^{\mathrm{ne}}}{\partial t^2} = 0 \qquad (1)^*$$

and by the material equations for the stationary and spatially-homogeneous media in the form

$$\mathbf{P}^{1}(\mathbf{r}, t) = \int_0^\infty dt_1 \int \hat{\varkappa}(\mathbf{r}_1, t_1)\mathbf{E}(\mathbf{r}-\mathbf{r}_1, t-t_1)d\mathbf{r}_1, \qquad (2)$$

$$\mathbf{P}^{\mathrm{ne}}(\mathbf{r}, t) = \iint_0^\infty dt_1 dt_2 \iint \hat{\chi}(\mathbf{r}_1, \mathbf{r}_2, t_1, t_2)\mathbf{E}(\mathbf{r}-\mathbf{r}_1, t-t_1) \times \mathbf{E}(\mathbf{r}-\mathbf{r}_1-\mathbf{r}_2, t-t_1-t_2)d\mathbf{r}_1 d\mathbf{r}_2 \ldots,$$

where $\hat{\kappa}$, $\hat{\chi}$, etc are tensors of the second, third, and higher ranks.

An exact solution of the system (1) and (2) is impossible, as is well known, even for unmodulated waves. Therefore, in order to consider problems with modulated waves, we shall use the method of slowly varying amplitudes; we confine ourselves first to relatively broad wave beams $a/\lambda \gg 1$ and extended wave packets $\tau_p/T \gg 1$. We can then obtain in lieu of the initial equation (1) approximate equations of the parabolic type, if we seek a field in the form

$$\mathbf{E}_n(t, \mathbf{r}) = \mathbf{e}_n A_n\left(\mu(\boldsymbol{\rho}_n\mathbf{r}), \sqrt{\mu}[\boldsymbol{\rho}_n\mathbf{r}], \sqrt{\mu}\left(t - \frac{\boldsymbol{\rho}_n\mathbf{r}}{u_n}\right)\right)\exp[i(\omega_n t - \mathbf{k}_n\mathbf{r})], \qquad (3)$$

Here $\mu \ll 1$—small parameter: different powers of μ preceding the different coordinates characterize the different degree of "slowness" of the spatial and temporal variables (see[4]); $\mathbf{e}_n$—polarization vector (we shall consider below problems in which the wave polarization is assumed to be constant), $\boldsymbol{\rho}_n$—unit vector parallel to the ray vector $\mathbf{s}_n = \partial\mathbf{k}_n/\partial\omega_n$, u_n—group velocity of the wave, $u_n^{-1} = |\mathbf{s}_n|$, $\mathbf{k}_n$—wave vector.

We substitute (3) in (1) and (2), following the usual procedure of the derivation of the abbreviated equations[3]. Retaining the terms of order μ, we get for the complex wave amplitude A_n the equation (we neglect the spatial dispersion of the medium)

$$[\mathbf{e}_n[\mathbf{k}_n\mathbf{e}_n]]\left\{\nabla - i\frac{1}{2}\boldsymbol{\rho}_n g_n\frac{\partial^2}{\partial\eta_n^2}\right\}A_n + i\frac{1}{2}\Delta_\perp A_n = F_n^{\mathrm{ne}}(A_m, A_l), \qquad (4)$$

where $\Delta_\perp$ is the Laplacian in the plane perpendicular to the vector $\boldsymbol{\rho}_n$ and

$$\eta_n = (u_n t - \boldsymbol{\rho}_n\mathbf{r}), \quad g_n = \partial^2 k_n/\partial\omega_n^2. \qquad (5)$$

In the nonlinear term F_n^{nl} we have also retained the quantities of order $\sim\mu$.

3. Before we proceed to consider specific problems, let us clarify the character of the effects connected with the different derivatives in (4). To this end, we consider the propagation of modulated waves in a linear medium and stop to analyze separately the spatial and temporal modulation of the wave. The propagation of the indicated waves in the medium is then described by the respective equations

$$\left\{\boldsymbol{\rho}_n\nabla - i\frac{1}{2}g_n\frac{\partial^2}{\partial\eta_n^2}\right\}A_n = 0, \qquad (6a)$$

$$\left\{\boldsymbol{\rho}_n\nabla + i\frac{1}{2k_n}\Delta_\perp\right\}A_n = 0. \qquad (6b)$$

The equations in (6) clarify the character of the approximations made on going over to them from Eq. (1). Analyzing the behavior of the Fourier components $\exp\{-i\Delta\mathbf{k}_n\cdot\mathbf{r}\}$ in (6b) and $\exp\{i[\Delta\omega_n\eta - \Delta\mathbf{k}_n\cdot\mathbf{r}]\}$ in (6a), we find that the real dependence of the wave vectors on the direction in an anisotropic medium is approximated by the parabolic relation

$$-2k_n(\Delta\mathbf{k}_n\boldsymbol{\rho}_n) = (\Delta k_{n,\perp})^2. \qquad (7a)$$

A parabolic relation describes in this approximation also the frequency dispersion of the medium

$$2(\Delta\mathbf{k}_n\boldsymbol{\rho}_n) = g_n(\Delta\omega_n)^2. \qquad (7b)$$

Comparison of (6a) and (6b) shows that the derivative with respect to the coordinate η_n in (6a) can be set in correspondence with the derivatives with respect to the coordinates in a plane perpendicular to the ray vector $\boldsymbol{\rho}_n$, with g_n corresponding to k_n^{-1}. If we confine ourselves to two-dimensional beams Eqs. (6) can be represented in a unified form:

$$\left\{\boldsymbol{\rho}_n\nabla + i\frac{1}{2}b_n\frac{\partial^2}{\partial\xi_n^2}\right\}A_n = 0. \qquad (8)$$

With this, we get in the temporal problem

$$b_n \to -g_n u_n^2, \quad \xi_n \to \eta_n; \qquad (8a)$$

and in the problem of the propagation of spatially-modulated waves

$$b_n \to k_n^{-1}, \quad \xi_n \to x_n. \qquad (8b)$$

If we put $b_n = 0$ in (8), then this equation has a solution in the form of traveling waves of arbitrary profile; their form is determined by the boundary conditions, which are specified at $z = 0$

$$A_n(\xi_n, z=0) = A_{n0}(\xi_n). \qquad (9)$$

In the analysis of the propagation of waves modulated in space, this case corresponds to the simplest variant of the geometrical-optics approximation, while for waves modulated in time it corresponds to the first

*$[\nabla[\nabla\mathbf{E}]] \equiv \nabla\times[\nabla\times\mathbf{E}]$

approximation of dispersion theory.

The parameter $b_n \neq 0$ is connected with the distortion of the profile of the traveling waves. In the case of the quasiplane wave (6b), the dispersion of the profile is due to diffraction ($k_n^{-1} \neq 0$), while for the quasi-monochromatic wave (6a) it is due to dispersion properties of the medium ($g_n \neq 0$). By solving (8) and assuming for simplicity a Gaussian initial amplitude profile of the wave

$$A_{n0}(\xi_n) = A_0 \exp\{-\xi_n^2/a^2\}, \tag{9a}$$

it can be shown that as the wave propagates in the medium the amplitude profile changes significantly over a length

$$z > l_b = {}^1/_2 a^2 b_n^{-1}. \tag{10}$$

In the spatial modulation problem $l_b = l_d = k_n a^2/2$ is the diffraction length of a beam of radius a having a plane phase front; in the temporal-modulation problem $l_b = l_s = \tau^2 (\partial^2 k/\partial\omega^2)^{-1}/2$ is the length of the dispersion spreading of the amplitude-modulated (AM) wave packet.

If phase modulation (converging or diverging beams, PM packets) is present besides the AM, then appreciable changes of the amplitude profile can occur over lengths shorter than l_b (l_d or respectively l_s). To describe the behavior of the signals experiencing AM and PM simultaneously, it is necessary to introduce the real amplitude A_n and the eikonal φ_n: $A_n \to A_n \exp\{-i\varphi_n/b_n\}$. Then for A_n and φ_n, choosing the z_n axis parallel to ρ_n, we get from (8)

$$\frac{\partial A_n}{\partial z_n} + \frac{\partial A_n}{\partial \xi_n}\frac{\partial \varphi_n}{\partial \xi_n} + \frac{1}{2} A_n \frac{\partial^2 \varphi_n}{\partial \xi_n^2} = 0, \tag{11a}$$

$$2\frac{\partial \varphi_n}{\partial z_n} + \left(\frac{\partial \varphi_n}{\partial \xi_n}\right)^2 = \frac{b_n^2}{A_n}\frac{\partial^2 A_n}{\partial \xi_n^2}) \tag{11b}$$

The case $b_n = 0$ ($g_n = 0$ or $k_n \to \infty$) corresponds to the geometrical-optics approximation. For a wave which is phase modulated at the input in accordance with the law

$$A_{n0}(\xi_n) = A_0(\xi_n) \exp\{i\xi_n^2/2b_n R\}, \tag{11c}$$

the amplitude profile experiences noticeable changes at $z_n = R$.

Summarizing the results presented in this section, we thus conclude that it is possible to trace a far-reaching space-time analogy in the behavior of broad wave beams and extended wave packets in a linear medium. In the succeeding sections we shall verify that this analogy can extend also to the nonlinear case.

2. CARRIER INTERACTION. GEOMETRICAL OPTICS AND FIRST APPROXIMATION OF DISPERSION THEORY

The simplest example of wave interaction of this type is the second-harmonic generation in a quadratic medium. In this case, the change of the wave amplitudes under conditions of exact phase synchronism is described by the equations

$$\left\{\rho_1 \nabla + i\frac{1}{2} b_1 \frac{\partial^2}{\partial \xi_1^2}\right\} A_1 = -i\sigma A_2 A_1^*,$$
$$\left\{\rho_2 \nabla + i\frac{1}{2} b_2 \frac{\partial^2}{\partial \xi_2^2}\right\} A_2 = -i\sigma A_1^2, \tag{12}$$

where σ—coefficient of nonlinear coupling. The indices 1 and 2 pertain to the waves with frequencies ω and 2ω. Each equation of (12) is written in its proper coordinates. As above, the influence of the temporal and spatial modulations of the waves on the course of the nonlinear processes will be analyzed separately. The equations (12) are applicable for both cases; the conversion to one of them is effected by the substitution (8a) or (8b) (compare with (8)).

In the case of normal incidence of the waves on the interface between the media, the system (12) assumes in the geometrical-optics approximation the form (we assume that $b_n = 0$ and also that the conditions of synchronism are satisfied for the case when the fundamental wave is ordinary and the harmonic extraordinary, and introduce a common coordinate system connected with the ray coordinates of the ordinary wave)

$$\frac{\partial}{\partial z} A_1 = -i\sigma A_2 A_1^*, \qquad \left(\frac{\partial}{\partial z} + \beta\frac{\partial}{\partial x}\right) A_2 = -i\sigma A_1^2 \tag{13}$$

for spatially modulated waves and

$$\frac{\partial}{\partial z} A_1 = -i\sigma A_2 A_1^*, \qquad \left(\frac{\partial}{\partial z} + \nu\frac{\partial}{\partial \eta}\right) A_2 = -i\sigma A_1^2 \tag{14}$$

for time-modulated waves.

In (13) and (14) $\eta = t - z/u_1$, $\beta = \widehat{\rho_1\rho_2}$—anisotropy angle, $\nu = (1/u_2 - 1/u_1)$ characterizes the difference between the group velocities.

We note that the system (14) can be obtained from (13) by setting definite parameters in the latter in correspondence with the analogous parameters in the temporal problem. Such parameters are x, β, and η, ν with

$$x \rightleftarrows \eta, \quad \beta \rightleftarrows \nu. \tag{15}$$

1. Generation of Second Harmonics by a Short Pulse (AM Signals)

For a wave modulated in amplitude only (a pulse or a bounded beam with a plane phase front), the harmonic-generation problem can be solved exactly[3)]. The solution is of considerable interest for the problem of frequency doubling of picosecond pulses; we shall therefore consider it in terms of the system (14). Going over in (14) to real amplitudes and phases $A_n \to A_n \exp(i\varphi_n)$ and putting for $z = 0$

$$A_1(z=0, \eta) = A_{10}(\eta), \quad A_2(z=0, \eta) = 0, \tag{16}$$

we can reduce the system (14) to the Ricatti equation

$$dA_2/dz + \sigma A_2^2 = \sigma A_{10}^2(\eta - \nu z). \tag{17}$$

Specifying the form of the pulse of the fundamental wave at the boundary

$$A_{10}(t) = A_0/\{1 + (t/\tau)^2\}, \tag{17a}$$

we arrive at the following solutions for A_1 and A_2 in an arbitrary section of the nonlinear medium:

3)We note that in the papers published to date this problem was solved only in the approximation using a given field of fundamental frequency.

$$A_1(\eta, z) = \frac{A_0}{(1+\tilde{\eta}^2)^{1/2}[1+(\tilde{\eta}-\tilde{z})^2]^{1/2}} \left\{ \operatorname{ch} y + \frac{\tilde{\eta}}{(l_k^2/l_\sigma^2-1)^{1/2}} \operatorname{sh} y \right\}^{-1}, \quad (18a)$$

$$A_2(\eta, z) = A_0 \frac{l_\sigma}{l_k} \frac{\tilde{z} \operatorname{ch} y + [(l_k^2/l_\sigma^2-1)^{1/2} - \tilde{\eta}(\tilde{\eta}-\tilde{z})(l_k^2/l_\sigma^2-1)^{-1/2}] \operatorname{sh} y}{[1+(\tilde{\eta}-\tilde{z})^2]\{\operatorname{ch} y + \tilde{\eta}(l_k^2/l_\sigma^2-1)^{-1/2} \operatorname{sh} y\}}, \quad (18b)$$

where

$$y = (l_\kappa^2/l_\sigma^2 - 1)^{1/2}[\operatorname{arc\,tg} \tilde{\eta} - \operatorname{arc\,tg}(\tilde{\eta} - \tilde{z})],$$

$$\tilde{\eta} = \eta/\tau, \quad \tilde{z} = z/l_\kappa.$$

In (18a, b) we have introduced the characteristic lengths:

$$l_\sigma = (\sigma A_0)^{-1} \quad (19)$$

—the so-called length of nonlinear interaction (70% of the conversion of the fundamental power into the harmonic takes place over this length in a non-modulated wave, see[3]) and

$$l_q = \tau/\nu \quad (20)$$

—the so-called quasistatic length—the length over which a noticeable spreading of the pulses of the fundamental radiation and of the harmonic takes place as a result of the group-delay effect.

Before we proceed to analyze the solutions (18), we note that a similar form will be assumed, evidently, by the solutions of the system (13); besides changing the variables, it is necessary here, obviously, to replace l_q by l_a:

$$l_a = a/\beta \quad (21)$$

—the so-called aperture length (see[5]).

The behavior of the amplitude profiles of the fundamental radiation and of the harmonic is determined by the ratio of the lengths l_σ and l_q (or else l_σ and l_a in the spatial problem). If

$$l_q > l_\sigma \quad (22)$$

then the amplitudes of the interacting waves change in exactly the same manner as for the unmodulated waves (compare with[3]); in this connection, we shall call the condition (22) quasistatic.

When $A_1 \gg A_2$ and $z \ll l_\sigma$ (specified field $A_1 = A_{10}(t - z/u_1)$) and $z < l_q$, the pulse of the harmonic is exactly the square of the fundamental pulse, and its amplitude increases in proportion to z, namely $A_2 = \sigma z A_{10}^2(t - z/u_1)$. When $z > l_q$, the harmonic pulse broadens; its duration increases in proportion to the length, $\tau_2 \approx \tau(1 + z/l_q)$, and the amplitude remains constant, $A_2 \approx \sigma l_q A_0^2$:[4] With increasing intensity of the fundamental radiation, so that $l_\sigma \ll l_q$, the frequency-doubling process becomes more and more quasistatic; the harmonic pulse broadens to a lesser degree[5].

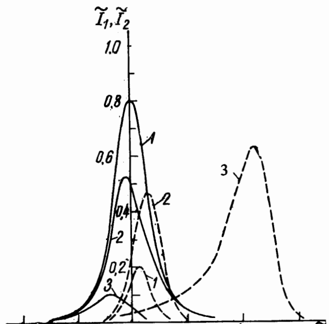

FIG. 1. Profiles of pulses of fundamental radiation $\tilde{I}_1 = A_1^2/A10^2$ (solid curves) and of the second harmonic $\tilde{I}_2 = A_2^2/A_{10}^2$ (dashed) for $(l_q/l\sigma)^2 = 1$ and different values of the traversed distance $\tilde{z} = z/l_q$: 1 – 0.5; 2 – 1.0; 3 – 5.0. The plots of the same figure also characterize the variation of the beam profiles in the case of generation of a harmonic by a beam with a plane phase front: $l_q \to l_a$, $\eta \to x = x/a$.

For the general case, Fig. 1 shows the variation of the wave profiles on going through the nonlinear medium. It also shows how the wave forms of the pulses of the fundamental radiation and the harmonic become modified as they propagate in the nonlinear medium. If the reaction of the harmonic on the fundamental radiation is appreciable, then the broadening of the harmonic pulse is accompanied by a certain narrowing of the fundamental pulse. At the same time, besides the shifting of the top of the harmonic pulse (in the coordinate system z, η) there can occur a noticeable shift of the peak of the fundamental radiation in the opposite direction (effects that are analogous to a certain degree to those occurring in laser amplifiers).

The curves of Fig. 2 illustrate the influence of the difference of the group velocities on the maximum efficiency of the frequency doubler; the shortening of the pulse duration (together with the associated shortening of the quasistatic length l_q) leads to a decrease in the efficiency of the doubler. For a KDP crystal at λ_1

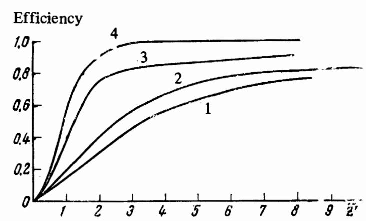

FIG. 2. Dependence of the energy coefficient of conversion of a frequency doubler (i.e., efficiency equal to W_2/W_{10}), excited by a laser pulse or by single mode radiation, on the reduced length $\tilde{z}' = z(l_q^{-1} + l_\sigma^{-1})$ for different values of $C = (l_q/l_\sigma)^2$: curve 1 – 0.5, 2 – 1.0, 3 – 10. For single-mode laser radiation $z' = z(l_a^{-1} + l_\sigma^{-1})$, $C = (l_a/l_\sigma)^2$. Curve 4 corresponds to conversion of plane monochromatic radiation.

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4) A similar result was obtained also in [6] in an analysis of frequency doubling of short laser pulses with synchronized modes.

5) We emphasize that the condition (22), $l_\sigma \ll l_q$, pertains to a nondissipative medium, and in a medium with appreciable losses the harmonic-generation process remains quasistatic at distances $l > l_q$ and at low intensities $l_\sigma \gg l_q$. In a specified pulsed field of fundamental radiation $A_1 = A_{10}(t - Z/u_1)\exp(-\delta_1 z)$ at a quasistatic length greatly exceeding the photon mean free path $l_q|\delta_2 - 2\delta_1| \gg 1$, the connection between the amplitudes of the fundamental wave and the harmonic wave becomes algebraic (and consequently the nonlinear response becomes quasistatic):

$$A_2 = -i\sigma A_{10}^2(t - z/u_n)[\exp(-2\delta_1 z) - \exp(-\delta_2 z)](\delta_2 - 2\delta_1)^{-1};$$

$n = 1, 2$. If $\delta_2 \gg \delta_1$, then $n = 1$, i.e., in this case the field of the harmonic that "breaks away" from the fundamental pulse attenuates rapidly; the shape of the harmonic pulse does not depend on the group delay. In the opposite case $\delta_1 \gg \delta_2$, $n = 2$ the effective generation of the harmonic occurs only in the first layers of the nonlinear medium ($z \lesssim \delta_1^{-1}$), where the process is still quasistatic.

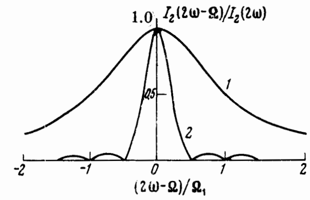

FIG. 3. Monochromatization of the frequency spectrum in frequency doubling under conditions when the group detuning differs from zero. 1 – spectrum of harmonic under conditions of quasistatic doubling; its width Ω_2 is of the order of the doubled width of the spectrum of the fundamental radiation Ω_1. Curve 2 – spectrum of harmonic produced at a distance $z > \pi/\nu\Omega_1$ at a nonzero group detuning (the curve was plotted for the case $\Delta = 0$, $\nu\Omega_1 z = 4\pi$).

$= 1.06\ \mu$ and $\tau_p \approx 10^{-12}$ sec, we get $l_q \approx 3$ cm; for $\lambda_1 = 0.53\ \mu$ and for the same duration we get $l_q \approx 0.3$ cm. Thus, the effects under consideration become decisive in cascade multipliers intended for the generation of picosecond pulses of ultraviolet radiation.

The foregoing results can be easily formulated also in spectral language. The formulas are particularly simple in the given-field approximation; assuming that not only the group difference but also the phase difference differ from zero for the average frequencies ω and 2ω, $\Delta = 2\omega c^{-1}[n(\omega) - n(2\omega)]$, we get

$$A_2(t, z) = -i\sigma \int_0^z A_{10}^2(t - z/u_2 + \nu y)\, e^{-i\Delta y} dy. \qquad (23)$$

Introducing the complex-amplitude spectra

$$S_n(\Omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} A_n(t)\, e^{-i\Omega t}\, dt, \qquad (24)$$

we get from (23)

$$S_2(\Omega, z) = -2i\sigma e^{i[(\Omega\nu-\Delta)/2-\Omega/u_2]z}\frac{\sin[\nu\Omega-\Delta]z/2}{[\nu\Omega-\Delta]}\int_{-\infty}^{+\infty} S_{10}(\Omega-\Omega')S_{10}(\Omega')\,d\Omega'. \qquad (25)$$

According to (25), the harmonic intensity spectrum $I_2(2\omega - \Omega) = S_2(\Omega) S_2^*(\Omega)$ becomes narrower with increasing path traversed in the nonlinear medium, in accordance with the formula

$$I_2(2\omega-\Omega) \sim \frac{\sin^2[\nu(2\omega-\Omega)-\Delta]z/2}{[\nu(2\omega-\Omega)-\Delta]^2}. \qquad (25a)$$

The maximum of the spectral density of the harmonic is reached at the frequency

$$\omega_{max} = 2\omega - \Delta/\nu. \qquad (25b)$$

At a fixed value of z, the width of the harmonic spectrum does not exceed $\Omega_{max} \approx 2\pi/z\nu$.

Formulas (25) thus show that the process of nonquasistatic $(z > l_q)$ frequency doubling of nonmonochromatic radiation can serve as the basis for the development of a sufficiently narrow-band generator with a smoothly variable frequency. Indeed, according to (25a), the spectrum of the harmonic can be much narrower than the spectrum of the fundamental beam (the foregoing is illustrated by Fig. 3); on the other hand, formula (25b) describes the "tuning characteristic" of such a generator. The output frequency of the harmonic generator can be varied by varying the phase difference $\Delta = 2\omega c^{-1}[n(\omega) - n(2\omega)]$, the temperature, or the orientation of the nonlinear crystal[6)].

The foregoing effects have simple spatial analogs—the narrowing of the angular spectrum of the harmonic in an anisotropic crystal with increasing crystal length, and the smooth variation of the angular frequency of the harmonic when the orientation of the anisotropic crystal relative to the diverging beam of the fundamental radiation is changed.

We note that if the fundamental beam has simultaneously sufficiently broad frequency and angular spectra, the frequency-doubling process can be used for the transformation of the frequency modulation into angular modulation.

2. Frequency Doubling of PM Signals; Shape of Harmonic Envelope

In the preceding subsection we analyzed the shape of the harmonic envelope only for an AM fundamental signal. Yet in many experimental situations interest attaches to the shape of the envelope of the harmonic in the case of a PM fundamental signal (strong PM is encountered in broadband optical signals produced in self-focusing liquids, see for example[8-11]). In the given-field mode and neglecting dispersion spreading, it is possible to use for the calculations the solution (23); in the more general case it is possible to use second order differential equations, for example such as (11). We shall not present here a general analysis (we note that the spectral formulas are valid for any type of modulation), and will confine ourselves to an important particular case. We assume that at the input to the nonlinear medium

$$A_{10}(t)\,\exp[i\varphi(t)] = A_0 \exp(-t^2/\tau^2 + i\gamma t^2),$$

and then we get for the intensity of the harmonic from (23), if $z < \tau/\nu$ and $(1/2)\,\gamma(\nu z)^2 < 1$,

$$I_2(t, z) = \sigma^2 I_1^2\left(t - \frac{z}{u_1}\right)\frac{\sin^2[t - z/u_1)\gamma\nu z]}{[(t - z/u_1)\gamma\nu z]^2}z^2, \qquad (26)$$

i.e., if the indicated conditions are satisfied, phase modulation of the fundamental radiation leads to a sharp amplitude modulation of the harmonic.

The AM period $T_M \approx \pi/\gamma\nu z$ decreases with increasing length of the nonlinear medium. Using (26), we can introduce the characteristic spatial scale

$$l_\varphi = \pi/\nu\,\frac{\partial\varphi}{\partial t}\Big|_{z=0} \qquad (26a)$$

which is the length that must be reached in order for the PM of the fundamental radiation to start influencing the form of the AM of the harmonic. The "phase" length l_φ has a simple intuitive analog—that is the so-called coherent length for a diverging beam in an anisotropic medium.

3. Shaping of Pulses in Frequency Doubling; Interaction of Pulses of Different Durations

One of the important applications of nonlinear wave effects is the shaping of ultrashort laser pulses; cer-

6) A laser of this type was realized recently by Carman and coworkers [7], who used as the fundamental radiation laser pulses passing through a self-focusing medium.

tain promises are offered in this sense by the use of frequency doubling. In a specified field, the shortening of the duration of a Gaussian pulse by approximately one half (see subsection 1 of this section) is attained under the conditions of quasistatic interaction ($z < l_q$); when $z > l_q$, the harmonic pulse broadens.

There is, however, another pulse-shaping mode, for which the group difference contributes to a narrowing of the harmonic pulse. We have in mind the interaction between a relatively narrow harmonic pulses, obtained from an external source, and a quasicontinuous fundamental radiation. Here, under conditions when the reaction of the harmonic on the fundamental radiation is appreciable in the presence of a group difference, the character of the energy exchange between the harmonic and the fundamental wave is different for the leading and the trailing edges of the pulse. Let us consider, for concreteness, the case $u_2 > u_1$ ($\nu < 0$); the leading edge of the harmonic pulse in this case "overtakes" the fundamental wave, acquiring an appreciable fraction of its energy; much less energy is left for the trailing part of the pulse.

A theory of this effect can be developed with the aid of the procedure described in subsection 1 of this section. An equation of type (17) can be written for

$$A_2(z=0,\eta) \neq 0, \quad \partial A_2(z,\eta)/\partial z|_{z=0} \neq 0;$$

its form is

$$\frac{dA_2}{dz} + \sigma A_2^2 = \sigma A_{10}^2(\eta - \nu z) + \sigma A_{20}^2(\eta - \nu z) - \nu\frac{\partial A_{20}(\eta - \nu z)}{\partial \eta} \tag{27}$$

Assume that at $z = 0$ and at the instant $t = 0$ a rectangular harmonic pulse (see Fig. 4) enters into a nonlinear medium excited by a quasicontinuous wave of fundamental frequency. According to (27), the shape of the harmonic pulse inside the medium changes; this change is described by a solution of the form

$$A_2(\eta, z) = A_0\,\mathrm{th}\,[F + \sigma A_0(z - \eta/\nu)], \tag{28}$$
$$\mathrm{th}\,F = \frac{A_{10}}{A_0}\,\mathrm{th}\left(\sigma A_0\frac{\eta}{\nu}\right) + \frac{A_{20}}{A_0}; \qquad A_0 = \sqrt{A_{10}^2 + A_{20}^2}.$$

On the leading front of the harmonic pulse ($t = z/u_2$, i.e., $z = \eta/\nu$), the amplitude experiences a jump equal to A_{20}, i.e., the same as at $z = 0$:

$$A_2(-0) = A_{10}\,\mathrm{th}\,(\sigma A_{10}z), \tag{29a}$$
$$A_2(+0) = A_{20} + A_{10}\,\mathrm{th}\,(\sigma A_{10}z). \tag{29b}$$

The foregoing is illustrated by Fig. 4; the width of the peak shaped near the leading front equals, according to (28), $\Delta\tau \approx \nu(\sigma A_{10})^{-1}$.

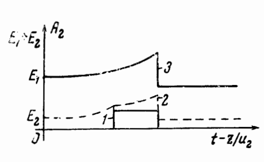

FIG. 4. Dynamics of the process of formation of an initially rectangular second-harmonic pulse of amplitude A_2 during the course of propagation in a field of quasicontinuous radiation of fundamental frequency (amplitude A_1). Plot 1 – form of the harmonic pulse at the input; 2 – after the leading front passes the distance $z < (\sigma A_{10})^{-1}$; 3 – stationary pulse of harmonic produced at $z > (\sigma A_{10})^{-1}$.

4. Three-Photon Parametric Interactions of Modulated Waves

Just as in the case of frequency doubling, we can separate here two sets of problems: parametric amplification in the field of a modulated pump (including pulsed), and parametric amplification of modulated waves in the field of a quasi-continuous pump. For the degenerate regime, the calculation must be based on the system (14); we shall henceforth call the wave frequency ω the signal ($A_1 \equiv A_s$), and the wave of frequency 2ω the pump ($A_2 \equiv A_p$).

In the given pump field ($A_p \gg A_s$), the intensity of the signal wave, according to (14), in a lossless system[7], is

$$I_s(t,z) = I_{s,0}(t - z/u_s)\exp\left\{2\sigma\int_0^z A_{p0}(t - z/u_c - \nu y)\,dy\right\}. \tag{30}$$

If, in particular, the pump is pulsed, then the signal acquires when $z \gg (\sigma A_{p,max})^{-1}$ a pulsed shape, regardless of the shape of the input signal. Practical interest attaches to the circumstance that under definite conditions the pulse duration of the signal can become shorter than the duration of the pump pulse; thus, an additional narrowing of picosecond laser pulses is possible.

Using (30), we can show that in the field of a Gaussian pump pulse of the form

$$A_{p0}(t) = A_0\exp\{-t^2/2\tau_p^2\}$$

at $\sigma A_0 z \gg 1$ and $\nu = 0$ (group synchronism), the signal pulse also becomes Gaussian, and its duration changes with increasing z in accordance with

$$\tau_s(z) \approx 1.4\tau_p(\sigma A_0 z)^{-1/2}. \tag{31a}$$

In real conditions, when $\sigma \neq 0$[8], the narrowing of the pulse takes place only for $z < \nu/\tau_p$, so that the limiting relative narrowing is of the order of

$$\frac{\tau_s^{(\lim)}}{\tau_p} \simeq \sqrt{\frac{\nu}{\sigma A_0\tau_p}}. \tag{31b}$$

When $z > \nu/\tau_p$, the signal pulse again broadens[9]. Thus, the qualitatively considered picture is similar to the picture of frequency doubling in a given field; however, the magnitude of the pulse narrowing for parametric interaction turns out to be more appreciable.

[7] The losses determine the threshold of the parametric amplification. A unique feature of parametric amplification in a modulated pump field is the fact that it is necessary to speak here not of a threshold power density, but of a threshold pump energy. From (14) we get for the reduced threshold energy ($W = \int I(t)dt$) the formula $W_p^{(thr)} = (1/2)\sqrt{\pi}(\delta/\sigma)^2\tau_p$, where δ is the damping decrement at the signal frequency.

[8] It should be noted, to be sure, that in birefringent crystals it becomes possible to create conditions under which both phase and group synchronism exist simultaneously.

[9] The effect of broadening of a weak parametric signal pulse in a pulsed pumping field at $z > l_q$ was considered also by Glenn [12]. It should also be noted that in nondegenerate parametric amplification, strong damping of one of the amplified waves causes the amplification process to become quasistatic also when $z > l_q$. The best example of such a situation is stimulated Mandel'shtam-Brillouin scattering, for which appreciable damping of the hypersonic wave is capable of "compensating" the group detuning $\nu = c^{-1}$ (see [13,14]).

Estimates show that in real situations it is possible to obtain $\tau_s(\lim)/\tau_p \approx 0.2$, whereas in quasistatic frequency doubling this quantity amounts to ~0.5–0.6. The behavior of a modulated signal in the field of quasicontinuous pumping depends on the ratio of the amplitudes A_s and A_p, and on the group difference ν.

In the degenerate regime, the spectrum of the amplified signal changes only when the reaction of the signal on the pump is appreciable. When $A_p \sim A_s$ and $\nu \neq 0$, an appreciable broadening of the signal spectrum (narrowing of the pulses) is possible. This effect, which is to a considerable degree similar to the process considered in subsection 4 of this section, makes it possible, as shown by estimates (see[15]) to obtain giant pulses (with a power exceeding the pump power) of parametric radiation.

In the nondegenerate regime ($\omega_s + \omega_d = \omega_p$), the picture becomes more complicated. The form of the spectra (or envelopes) of the modulated waves at frequencies ω_s and ω_d in the quasi-continuous pumping field changes in the given pumping field if the group velocities of the signal and different frequency waves are different, ($\nu = 1/u_s - 1/u_d \neq 0$).

Solving the system of corresponding linear abbreviated equations

$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_s}\frac{\partial}{\partial}\right\} A_s+\delta_s A_s=-i\sigma_s A_{p0} A_d^* e^{i\Delta z}, \tag{32a}$$

$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_d}\frac{\partial}{\partial t}\right\} A_d+\delta_d A_d=-i\sigma_d A_{p0} A_s^* e^{i\Delta z}, \tag{32b}$$

we can readily show that in the region where the pump greatly exceeds threshold the width of the spectrum of the amplified signal

$$\Delta\Omega \approx \frac{4.5}{|\nu|}|A_{p0}|\sqrt{\frac{2\sigma_s\sigma_d}{z}} \tag{32c}$$

decreases monotonically with increasing distance.

An analysis of the nondegenerate amplification with allowance for the reaction on the pump at $u_s \neq u_d$ is a rather complicated problem. From the point of view of narrowing down the pulses, particular interest attaches to the case of essentially unequal damping decrements at the frequencies ω_s and ω_d.

3. ENVELOPE INTERACTION; CROSS MODULATION OF MODULATED WAVES IN A MEDIUM WITH POLARIZATION THAT IS CUBIC IN THE FIELD. GEOMETRICAL ANALYSIS

By way of an example of nonlinear interactions of modulated waves in a medium with nonlinear polarization in the form $P^{nl} = \Theta E^3$, we consider the interaction of wave packets

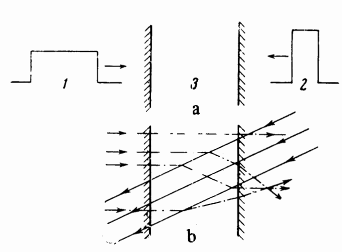

FIG. 5. a – interaction of pulses 1 and 2 in a cubic medium 3; b – spatial analog of Fig. a – interaction of bounded beams of weak wave of frequency ω_1 are shown dashed; rays of intense wave of frequency ω_2 – solid.

$$E_1(t, z)=A_1(t, z)\exp[i(\omega_1 t-k_1 z)],$$
$$E_2(t, z)=A_2(t, z)\exp[i(\omega_2 t+k_2 z)],$$

propagating against each other in a nonlinear medium of dimension L (see Fig. 5). Assuming that the frequencies ω_1 and ω_2 are noncommensurate, we obtain for the complex amplitude A_1 and A_2 abbreviated equations in the form

$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_1}\frac{\partial}{\partial t}\right\} A_1 = -i\{\sigma_{11}|A_1|^2+\sigma_{12}|A_2|^2\}A_1, \tag{33}$$

$$\left\{\frac{\partial}{\partial z}-\frac{1}{u_2}\frac{\partial}{\partial t}\right\} A_2 = -i\{\sigma_{21}|A_1|^2+\sigma_{22}|A_2|^2\}A_2, \tag{33b}$$

In (33)

$$\sigma_{mn}=\frac{\omega_m^2}{k_m}\frac{2\pi}{c^2}\Theta(\omega_m+\omega_n-\omega_n).$$

The system (33) should be solved with the boundary conditions

$$A_1(t, z=0)=A_{10}(t), \quad A_2(t, z=l)=A_{20}(t). \tag{33'}$$

Eqs. (33) can be solved in general form; for example, for A_1 we have

$$A_1(t, z)=A_{10}(t-z/u_1)\exp\{-i\sigma_{11}|A_{10}(t-z/u_1)|^2-i\varphi_1(t, z, L)\}, \tag{34a}$$

where

$$\varphi_{1a}(t, z, L)=-\sigma_{12}\int_0^z |A_{20}(t-\nu_+ z+\nu_+ y)|^2 dy, \tag{34b}$$
$$\nu_+ = u_2^{-1}+u_1^{-1}.$$

A_2 is expressed similarly.

Thus, in a cubic medium, the modulated waves can exhibit not only self-action but also an influence on the phase of the other waves propagating in the medium (cross modulation). Let us analyze the behavior of the additional phase φ_{1a} (34b) due to the cross-modulation effect. Let us assume that the amplitude $A_{20}(t)$ is harmonically modulated, $A_{20}(t) = A_0 \cos \Omega t$; we then obtain

$$\varphi_{1a}=-\frac{1}{2}\sigma_{12}A_0^2\left[z+\frac{\sin(\Omega\nu_+ z)}{\Omega\nu_+}\right]\cos\Omega\left(t-\nu_+ L+\frac{1}{2}\nu_+ z\right). \tag{35}$$

The index of the phase modulation of the wave increases linearly with distance if

$$\Omega\nu_+ z < \pi/2. \tag{36}$$

If the interacting waves propagate in identical directions, it is necessary to write in (36) $\nu = u_2^{-1} - u_1^{-1}$ in lieu of ν_+. Particular interest attaches to an analysis of the phase φ_{1a} in a pulsed field

$$|A_{20}(t)|^2 = I_{20}\{1(t)-1(t-\tau)\} \tag{37}$$

($1(t)$ is the Heaviside unit step function and τ is the pulse duration), which increases[10)] in a nonlinear

10)A case of practical interest, corresponding to such a formulation of the problem, is the problem of cross modulation of long ($\tau_p \approx 10^{-8}$ sec) SRS pulses propagating in the direction of the scattered radiation, and ultrashort SRS pulses ($\tau_p \approx 10^{-11}$ sec) propagating at 180° (see [10]).

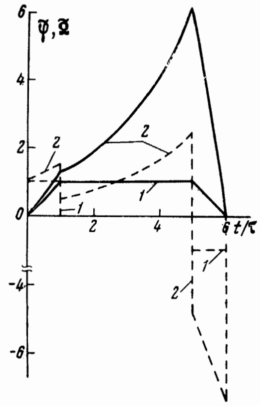

FIG. 6. Plots of the reduced phases $\tilde{\varphi} = K\varphi_{1a}(K = -\nu_+/\sigma_{12} I_{20})$ (solid curves) and of the reduced deviation of the carrier frequency $\tilde{\Omega} = K\, \partial\varphi_{1a}/\partial t$ (dashed) at the different values of α : $1 - \alpha = 0$, $2 - \alpha = 0.2\, \nu_+/\tau(\nu_+ L = 5\tau)$.

medium. It is necessary to add under the integral in (34) a factor $\exp[2\alpha(z - y)]$, where α is the growth coefficient. In this case the change of the phase at the exit from the medium, z = L, is determined by the expression

$$\varphi_{1a}\left(-\frac{2\alpha}{\sigma_{12}I_{20}}\right)=\begin{cases}0, & t\leqslant 0,\ t\geqslant\tau+\nu_+L\\ e^{2\alpha t/\nu_+}-1 & 0\leqslant t\leqslant\tau\\ e^{2\alpha t/\nu_+}[1-e^{-2\alpha\tau/\nu_+}], & \tau\leqslant t\leqslant\nu_+L.\\ e^{2\alpha L}-e^{2\alpha(t-\tau)/\nu_+} & \nu_+L\leqslant t\leqslant\tau+\nu_+L\end{cases} \quad (38)$$

A plot of φ_{1a} (38) is shown in Fig. 6. The same figure shows the behavior of the frequency deviation of the signal wave, $\Omega = d\varphi_{1d}/dt$[11]. Thus, owing to the change in the carrier frequency, the spectrum of the wave can broaden. In the analysis of the phase φ_{1a} it was tacitly assumed that the signal wave is monochromatic. In the case of a pulse signal of frequency ω_1, it is possible to determine the character of the change of the phase φ_{1a} by plotting along the abscissa axis the times of appearance of the leading and trailing fronts of the signal, the time being reckoned from the instant (t = 0) of the appearance of a pulse of frequency ω_2 on the boundary of the nonlinear medium z = L; the time interval determines the change of the phase φ_{1a}.

From the analysis of the spectrum connected with the phase (38) we can deduce one important circumstance: the signal spectrum has an asymmetrical distribution, whereby in a focusing medium $(\sigma_{21} > 0)$ the spectrum decreases more slowly in the low-frequency region, and in a defocusing medium $(\sigma_{12} < 0)$ in the region of high frequencies.

A spatial analog of the just investigated problem is the interaction of bounded beams, for which the foregoing results are valid, including the plots of Fig. 6 $(t \to x)$. Inasmuch as the beam is phase modulated, a plane-parallel beam becomes diverging (see Fig. 5). If the beam under consideration has a definite initial divergence, then in the field of an intense wave the cross modulation may increase or decrease its divergence. The latter effect depends on the sign of the nonlinearity of the medium and on the boundary near which the beam interaction takes place.

[11]Under real conditions, the finite rise time of the fronts of the pulse $|A_{20}(t)|^2$ leads to a continuous change of frequency.

4. WAVE OPTICS AND SECOND APPROXIMATION OF DISPERSION THEORY

1. Frequency Doubling in the Presence of Dispersion Spreading of Wave Packets

We now proceed to an analysis of nonlinear interaction of wave packets with allowance for second derivatives in the equations (4). We are thus dealing with allowance for the dispersion spreading of the packets. It should be borne in mind that a consideration of these effects becomes significant in practice when $\tau_p \approx 10^{-13}$ sec, since in typical nonlinear crystals we have $\partial^2 k/\partial\omega^2 \approx 10^{-27}$ sec^2/cm, and consequently, when $\tau \le 10^{-13}$ sec we have $l_d \le 10$ cm. We turn to an investigation of the frequency doubling process. The effects of dispersion spreading take part here, as can be readily verified, as an appreciable change of the form of the spectrum of the harmonic and in a change of the rate of growth of the energy of the harmonic with increasing distance. In the given-field approximation, this process is described by the equations

$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_1}\frac{\partial}{\partial t}-i\frac{1}{2}g_1\frac{\partial^2}{\partial t^2}\right\}A_1=0, \quad (39)$$
$$\left\{\frac{\partial}{\partial z}+\frac{1}{u_2}\frac{\partial}{\partial t}-i\frac{1}{2}g_2\frac{\partial^2}{\partial t^2}\right\}A_2=-i\sigma A_1^2e^{-i\Delta z}$$

with boundary conditions of the type (16).

The solution of the system (39) leads to the following expression for the frequency spectrum of the harmonic:

$$S_2(\Omega,z)=i\sigma e^{-i\psi_2(\Omega)z}\int\int_{-\infty}^{+\infty}S_{10}(\Omega_1')S_{10}(\Omega_1'')e^{-i\psi z/2} \times\frac{\sin(\psi z/2)}{\psi/2}\delta(\Omega-\Omega_1'-\Omega_1'')d\Omega_1'd\Omega_1''. \quad (40)$$

Here

$$\psi(\Omega,\Omega_1',\Omega_1'')=\Delta+\psi_1(\Omega_1')+\psi_1(\Omega_1'')-\psi_2(\Omega),$$
$$\psi_n(\Omega)=\Omega/u_n+{}^1/_2g_n\Omega^2.$$

An appreciable contribution to the spectrum $S_2(\Omega, z)$ is made by waves whose spectral components satisfy the relations

$$\Omega=\Omega_1'+\Omega_1''; \quad \psi(\Omega,\Omega_1',\Omega_1'')=0, \quad (41)$$

and the latter relation, taking into account (7b) and the expression for Δ, can be written in the form

$$k_1(\omega+\Omega_1')+k_1(\omega+\Omega_1'')=k_2(2\omega+\Omega). \quad (42)$$

Thus, it turns out that an appreciable contribution to the spectrum of the harmonic is made by spectral components of the fundamental wave, for which the phase synchronism condition is satisfied. The presence in the general case of an aggregate of synchronous interactions can lead to a modification of the spectrum of the harmonic.

If $|g_1| \le |g_2|$, then there can exist only two synchronous interactions; in this case

$$\psi'=\Delta-\nu\Omega-{}^1/_2g_2\Omega^2,$$

and the spectrum of the harmonic

$$S_2(\Omega;z)=i\sigma\exp\left\{-i\psi_2(\Omega)z-i\psi'(\Omega)\frac{z}{2}\right\}\frac{\sin(\psi' z/2)}{\psi'/2}$$

$$\times \int_{-\infty}^{+\infty} S_{10}(\Omega-\Omega_1')S_{10}(\Omega_1')\,d\Omega_1' \qquad (43)$$

has zero values that are non-equidistantly spaced (see (25)).

Expression (43) makes it possible to analyze harmonic generation with various types of modulation of the fundamental.

Integrating the solutions of (39) with respect to time, we can readily see that the character of the dependence of the harmonic pulse energy on the distance z covered in the nonlinear medium depends essentially on the relation of z with $l_{p_n} = \tau^2/(2\partial^2 k_n/\partial\omega_n^2)$. The corresponding formulas are particularly clear in the case of $\nu = 0$ (under the conditions of group synchronism).

For the case $l_{d1} = l_{d2}/2 = l_d > z$ we have

$$W_2(z) = \frac{4\pi}{cn}\sqrt{\frac{2}{\pi}}\sigma^2\frac{W_1^2}{\tau}z^2, \qquad (44)$$

For $z > l_d$ we get

$$W_2(z) = \frac{16\pi}{cn}\sqrt{\frac{2}{\pi}}\sigma^2\frac{W_1^2}{\tau}l_d z. \qquad (45)$$

We note, finally, that by using the foregoing formulas we can determine the optimal pulse duration of the fundamental radiation having a fixed energy, i.e., the duration at which the efficiency of the frequency doubler of length z is maximal. This problem is analogous to the problem of optimal spatial focusing in a frequency doubler (it can therefore be called the problem of optimal compression).

An analysis of the solutions (39) leads to a physically lucid result: the optimal pulse has a duration

$$\tau = \tau_0 \approx \sqrt{z\frac{\partial^2 k_1}{\partial\omega^2}}.$$

Of course, the problems of optimal compression occur also for other nonlinear processes (parametric amplification, multiphoton effects, etc.).

2. Influence of Finite Pulse Duration on the Behavior of a Bounded Light Beam--Nonstationary Diffraction

The theory developed above for nonstationary effects pertains to interactions between plane wave packets. Allowance for a simultaneously present spatial modulation (bounded beams) entails no great difficulty if the characteristic spatial dimensions l_q and l_a (see (20) and (21) and l_d and l_s (see (10)) differ greatly in magnitude.

The problem becomes more complicated if $l_q \sim l_a$ and $l_d \sim l_s$. Then the effects of temporal and spatial modulation appear simultaneously and can greatly influence each other.

Thus, divergence of the fundamental beam (angle α_1) can greatly influence the temporal and spectral characteristics of a frequency doubler for picosecond pulses. It is easy to verify that if $\alpha_1 \approx \Omega_1 d\Theta/d\omega_1$, where the derivative $d\Theta/d\omega$ characterizes the sensitivity of the synchronism angle to the frequency, and Ω_1 is the width of the fundamental-radiation spectrum, there will be no monochromatization of the beam as a whole (of the type described in subsection 1 of Sec. 2), even when $z > l_q = \tau/\nu$. On the other hand, an analysis shows that in many cases the limiting parameters of "giant" parametric pulses are determined by the spatial limitation on the beam.

An analysis of these problems is, however, beyond the scope of the present article. We therefore confine ourselves here to a brief discussion of the linear problem, pertaining to the indicated circle of problems, namely the problem of the diffraction of a short pulse in a linear non-dispersive medium. The initial equation is in this case a second-order equation in the form

$$\frac{\partial A}{\partial z} = -i\frac{1}{2k}\Delta_\perp A + \frac{i}{\omega}\frac{\partial^2 A}{\partial z\,\partial\eta}. \qquad (46)$$

The notation in (46) is standard (see Sec. 2); the influence of the temporal modulation on the diffraction is described by a mixed derivative. Using (46) for the spectral amplitude $S(\Omega)$

$$A(t) = \int_{-\infty}^{+\infty} S(\Omega)e^{i\Omega t}\,dt$$

of the frequency spectrum of the packet, we obtain a parabolic equation with a diffusion coefficient that depends on Ω:

$$\frac{\partial S(\Omega)}{\partial z} = \frac{1}{2ik(1+\Omega/\omega)}\Delta_\perp S(\Omega). \qquad (47)$$

From (47) it follows directly that the frequency spectrum, and consequently also the form of the wave packet on the beam axis is deformed with increasing z; the short-wave components are diffracted more slowly than the long-wave components; the indicated effect is noticeable at not too small Ω/ω, i.e., for pulses of duration of several optical periods.

5. CONCLUSION

The foregoing results thus show that the use of approximate parabolic equations makes it possible to solve an extensive group of problems connected with non-stationary interactions of light waves. Among the most important parameters occurring in nonstationary nonlinear optics is the group detuning ν; such processes as generation of harmonics by short pulses and parametric amplification in a pulsed pumping field proceed effectively so long as $z < l_q$ (the effect of dispersion spreading can be neglected in real situations up to $\tau \approx 10^{-13}$ sec, since usually $\partial^2 k/\partial\omega^2 \approx 10^{-27}$ sec^2/cm). In this connection, the problem of finding media in which the conditions of phase and group synchronism are simultaneously satisfied becomes important[12)]. In a KDP crystal, at $\lambda_1 \approx 1.06\ \mu$, the directions of the indicated synchronisms differ somewhat; they can apparently be aligned by changing the crystal parameters.

At the same time, as already indicated, there is another important class of nonstationary wave phenomena, for which the group detuning increases the efficiency. We have in mind three-photon interactions of optical signals with essentially differing durations, for example parametric amplification of a short pulse

12)Obviously we are referring to temporal analogs of media that admit of synchronous interactions without the diaphragm aperture effect (see [8]).

in a quasi-continuous pumping field. An examination of the nonstationary wave phenomena, from the point of view of the formation of ultrashort pulses, was reported by us in[15]. To understand many features of nonlinear interactions of ultrashort pulses and to systematize the theoretical results, the space-time analogy in the theory of nonlinear interactions of modulated waves, investigated in detail above, is very useful.

We note finally that an investigation of nonstationary wave phenomena can be a useful method of analyzing the amplitude-phase structure of broadband optical signals, particularly broadband signals obtained in self-focusing liquids (see[8,9,15,16]. As shown in Sec. 2, at an essentially nonquasistatic frequency doubling ($z > l_q$), the form of the envelope of the harmonic turns out to be very sensitive to the form of the modulation of the fundamental radiation (AM, PM); certain information can be obtained also by measuring the fine structure of the harmonic spectrum.

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