

É. A. Ibragimov and T. S. Usmanov

*Institute of Electronics, Academy of Sciences of the Uzbek SSR, Tashkent*

(Submitted 15 August 1983)

Zh. Eksp. Teor. Fiz. **86**, 1618–1631 (May 1984)

The given-field approximation (GFA) and the recently developed given-intensity approximation (GIT) are widely used in the theory of nonlinear waves. A new approximation is developed in the present paper. This strong-interaction approximation (SIA) takes into account strong energy transfer between interacting modulated waves in anisotropic dispersive media. It is shown that the range of validity of SIA is considerably greater than that of GFA or GIA. In this paper, we use SIA to consider the stationary interaction between real wave beams in homogeneous and inhomogeneous nonlinear media, and the nonstationary interaction between waves. The effect of cubic nonlinearity on frequency multiplication in the presence of a strong pump field is examined. An expression is obtained for the limiting efficiency of conversion of pump energy into the energy of generated wave under realistic conditions. Several effects that are absent from GFA and GIA are identified. They include the dependence of the angular dispersion parameter on the pump power density and the shortening of the harmonic pulse under nonstationary excitation by a phase-modulated pump pulse.

## INTRODUCTION

The given-field approximation<sup>1-3</sup> (GFA) has been widely used for a considerable time in the theory of nonlinear wave interaction in dispersive media. The GFA approach it to take the complex amplitude of the original wave as given, i.e., to ignore the reaction of the generated or amplified waves on the pump wave. This provides a correct description but only for weakly-interacting waves. On the other hand, the nonlinear wave equation with allowance for reaction has been solved only for special cases that are not close enough to real beams in real nonlinear media.<sup>4-6</sup> The given-intensity approximation<sup>7-9</sup> (GIA) has recently been developed for nonlinear wave interactions. In contrast to GFA, this does not impose any restrictions on the phase of the pump wave, and the pump-wave intensity is regarded as given. GIA provides a satisfactory description of waves in nonlinear media, but only on the scale of one nonlinear interaction length.<sup>7</sup> In other words, this approximation is not valid for strongly interacting waves. Numerical methods have now been developed for the solution of nonlinear wave equations, which are capable of describing the interaction of focused beams or waves in inhomogeneous nonlinear media.<sup>10,11</sup> However, it is very desirable to develop analytical methods capable of describing strong interactions between waves, and yielding manageable results.

In this paper, we develop the strong-interaction approximation (SIA) for the analysis of nonlinear interactions between waves in dispersive media. SIA differs from GFA and GIA in that it imposes no restriction on either the phase or the amplitude of the interacting waves. The new approximation is founded on the physical principle that strong energy transfer between waves in weakly-dispersive media can occur only between almost plane, i.e., ideal, waves. The mathematical foundation of this approximation is the method of successive approximations. The known<sup>4</sup> plane-wave solution of the equations is used as the zero-order approxi-

mation, which means that the degree of approximation is measured by the difference between the real and the plane waves. The method is valid for both weak and strong interactions between real waves in real media that lead to complete transfer of energy from the original wave to the resulting wave. Several problems in nonlinear optics are analyzed below on the basis of SIA.

## 1. FUNDAMENTALS OF THE METHOD

Let us consider the principle of solving nonlinear wave equations that describe harmonic generation in SIA. In its most general form, the process of degenerate triple-frequency interaction is described by the following set of equations:

$$\begin{aligned} \frac{\partial A_1}{\partial z} + \alpha f_1(A_1, A_2) &= -i\gamma A_1^* A_2 e^{i\Delta h z}, \\ \frac{\partial A_2}{\partial z} + \alpha f_2(A_1, A_2) &= -i\gamma A_1^2 e^{-i\Delta h z}, \end{aligned} \quad (1.1)$$

where  $A_1$  and  $A_2$  are complex amplitudes of the fundamental and the second harmonic, respectively,  $\gamma$  is the nonlinear coupling coefficient,  $\Delta k = 2k_1 - k_2$  is the phase mismatch, and  $\alpha$  is a dimensionless constant. When  $\alpha = 0$ , the above equations describe the interaction of unmodulated plane waves in a nonlinear medium, and their solution is known.<sup>4</sup> It is clear that the simple plane-wave theory is inadequate for the correct description of nonlinear processes in real situations. The spatial and temporal modulation of real laser radiation limits the wave interaction efficiency in real nonlinear media, and the functions  $f_1(A_1, A_2)$  and  $f_2(A_1, A_2)$  in (1.1) describe the influence of factors restricting this efficiency. One or other of these factors becomes dominant, depending on particular experimental conditions.

The solution of (1.1) will be found for the most general case, so that, for the moment, the form of the functions will not be specified. The method put forward below can be used

to obtain solutions of (1.1) that are close to the exact solutions and are valid for a broad range of functions describing both weak and strong energy transfer between the waves. We shall seek the solution in the form of an expansion in powers of  $\beta = \alpha(l_{nl}/a)$ :

$$A_1 = \sum_{n=0}^{\infty} \beta^n A_{1,n} = \sum_{n=0}^{\infty} \beta^n (u_{1,n} + iu_{2,n}) e^{i\varphi}, \quad (1.2)$$

$$A_2 = \sum_{n=0}^{\infty} \beta^n A_{2,n} e^{-i\pi/2} = \sum_{n=0}^{\infty} \beta^n (v_{1,n} + iv_{2,n}) e^{i(2\varphi - \pi/2)}$$

where  $l_{nl} = \gamma^{-1} A_0^{-1}$  is the nonlinear interaction length and  $a$  is the width of the beam (or the pulse length, depending on the particular special case). The function  $\varphi$  does not depend on  $z$  and is found from the boundary conditions. The solution of zero order in  $\beta$  is the known exact plane-wave solution.<sup>4</sup> For second-harmonic generation, the boundary conditions at  $z = 0$  are:

$$A_{1,0} = c(x, y, t) e^{i\varphi(x, y, t)} \quad (1.3)$$

$$A_{1,n} = 0 \quad (n \neq 0); \quad A_{2,n} = 0,$$

where  $c$  and  $\varphi$  are, respectively, the amplitude and phase distributions at entry to the nonlinear medium. Substitution of the solutions in the form of (1.2) into (1.1) gives a set of linear differential equations for each quartet of terms of order  $n$ , and the solutions of this set are the functions

$$u_{1,n} = \frac{1}{u_{1,0}} (Q_{1,n} - v_{2,0} v_{1,n}), \quad u_{2,n} = u_{1,0} D_{1,n},$$

$$v_{1,n} = -u_{1,0} D_{2,n}, \quad v_{2,n} = \frac{2}{u_{1,0}} (Q_{2,n} + v_{2,0} u_{2,n}), \quad (1.4)$$

where  $D_i, Q_i$  involve terms of order  $n - 1$  or lower:

$$D_p = \int_0^z \frac{dz}{u_{1,0}^2} (2Q_p + F_p),$$

$$Q_1 = \int_0^z (v_{2,0} - u_{1,0} F_3) dz,$$

$$Q_2 = \frac{1}{2} \int_0^z (u_{1,0} F_4 + 2v_{2,0} F_2) dz,$$

$$F_1 = -\beta \operatorname{Re} G_1 + \sum_{k=1}^{n-1} (u_{1,k} u_{1,n-k} - u_{2,k} u_{2,n-k}),$$

$$F_2 = -\frac{\beta}{u_{1,0}} \operatorname{Im} G_2 + \frac{1}{u_{1,0}} \sum_{k=1}^{n-1} (u_{1,k} v_{2,n-k} - u_{2,k} v_{1,n-k}),$$

$$F_3 = -\beta \operatorname{Re} G_2 + \sum_{k=1}^{n-1} (u_{1,k} u_{1,n-k} - u_{2,k} u_{2,n-k}),$$

$$F_4 = -\beta \operatorname{Im} G_1 + \sum_{k=1}^{n-1} (u_{2,k} u_{1,n-k} + u_{1,k} u_{2,n-k}), \quad (1.5)$$

where  $G_q = e^{-iq\varphi} f_q(A_q)$  and the sums on the right-hand sides of the expressions for  $F$  vanish for  $n = 1$ .

Under certain particular conditions, namely, when the phase difference between the first and second harmonics is constant, we can show, using the estimates given in Ref. 12, that the difference between first-order SIA and the exact solution does not exceed 5% for  $\beta < 1$ .

For a rigorous demonstration of the validity of (1.4), we would have, generally speaking, to prove the uniform convergence of the series (1.2) but, as will be shown below, comparison with numerical calculations indicates that very good agreement between exact and approximate solutions is achieved provided  $\beta$  is not too large. In the ensuing calculations of conversion efficiency and other quantities, we shall confine our attention to terms of the second order in small quantities. This turns out to be quite sufficient for the solution of all the problems that we shall consider.

## 2. NONLINEAR WAVE INTERACTION IN THE ABSENCE OF EXACT PHASE MATCHING

The efficacy of SIA is most simply illustrated in the case where nonlinear interaction is restricted by the fact that the phase matching conditions are not satisfied. We then have  $f_1 = 0, \alpha f_2(A_1, A_2) = i\Delta K A_2$ , so that

$$\frac{dA_1}{dz} = -i\gamma A_1^* A_2, \quad \frac{dA_2}{dz} + i\Delta K A_2 = -i\gamma A_1^2. \quad (2.1)$$

Exact solutions of (2.1) are well known<sup>4</sup> and can be compared with the SIA solutions. The conversion coefficient  $\eta(z)$  that represents energy transfer between the waves is given by the following expression in terms of the first- and second-order approximations:

$$\eta(z) = v_{2,0}^2 - \Delta s^2 [v_{1,1}^2 + 2v_{2,0} v_{1,2} + v_{2,1}^2], \quad (2.2)$$

where  $\Delta s = \Delta k l_{nl}$  is the normalized mismatch.

Substituting for  $v_{i,j}$ , which are the solutions of (2.1), into (2.2) and using the SIA method, we obtain the following expression from (1.4) and (1.5):

$$\eta(z) = \operatorname{th}^2 z - \frac{\Delta s^2}{32} \frac{\operatorname{sh} z}{\operatorname{ch}^3 z} \left[ \frac{\operatorname{sh}(4z)}{4} - z \right]. \quad (2.3)$$

The  $z$  coordinate is also normalized to  $l_{nl}$ . As expected, (2.3) does not contain terms proportional to  $\Delta s$ . This is a direct reflection of the fact that  $\eta(z)$  should not depend on the sign of the phase mismatch. It follows that the next-order non-zero terms in the expansion given by (2.3) must be proportional to  $\Delta s^4$ , which enhances the precision of SIA. Comparison of (2.3) with the exact solution shows that, for  $\Delta s < 1$ , the two results agree to a high degree of precision up to the values of  $z$  for which the conversion process begins to occur in the reverse direction. This is not unexpected since the expansion up to the second order in  $\beta$  can describe the conversion process only up to the first maximum. However, SIA can be substantially improved by exploiting the fact that, after the first maximum has been reached, the quantity  $\eta$  can be obtained by symmetric reflection at the point at which the maximum was reached. It is, in fact, readily verified that this problem is symmetric relative to the point  $\xi_{\max}$  at which the

phase changes sign. From Ref. 4, we have

$$\xi_{\max} = \frac{1}{v_b} \int_0^{\pi/2} \frac{dt}{(1-v_b^4 \sin^2 t)^{1/2}}, \quad (2.4)$$

where  $v_b$  is the amplitude of the second wave at the point of maximum:

$$v_b^2 = \left\{ \frac{\Delta s}{4} + \left[ 1 + \left( \frac{\Delta s}{4} \right)^2 \right]^{1/2} \right\}. \quad (2.5)$$

Solutions obtained with the aid of (2.3) using reflection at  $\xi_{\max}$  are shown in Fig. 1. It is clear that SIA gives sufficient precision even when  $\Delta s$  is large and the maximum conversion coefficient is much less than 50%.

### 3. HARMONIC GENERATION IN ANISOTROPIC MEDIA

The general form of equations for the triple-frequency interaction of waves in an anisotropic medium with quadratic nonlinearity is

$$\begin{aligned} \frac{\partial A_1}{\partial z} + \beta_1 \frac{\partial A_1}{\partial x} &= -i\gamma_1 A_2^* A_3 e^{-i\Delta k z}, \\ \frac{\partial A_2}{\partial z} + \beta_2 \frac{\partial A_2}{\partial x} &= -i\gamma_2 A_1^* A_3 e^{-i\Delta k z}, \\ \frac{\partial A_3}{\partial z} + \beta_3 \frac{\partial A_3}{\partial x} &= -i\gamma_3 A_1 A_2 e^{i\Delta k z}, \end{aligned} \quad (3.1)$$

where the  $x$  axis lies in the principal optical plane of the crystal, at right angles to the direction of propagation of the beam,  $\beta_i$  are the birefringence angles for the  $i$ th wave, and  $\Delta k = k_1 + k_2 - k_3$  is the wavenumber mismatch.

It is well known that anisotropy of the medium restricts conversion efficiency and leads to a slower growth of the second harmonic as compared with the plane-wave case<sup>5,13</sup> because of the so-called aperture effects. The influence of these effects is usually estimated by means of the following characteristic spatial scales: the aperture length  $l_a = a/\beta$  and the coherence length of the divergent beam,<sup>5</sup> or the dispersion length,  $l_\gamma = 2\pi/\beta K\theta$ , where  $\theta$  is the beam divergence angle at fundamental frequency and  $a$  is its aperture. We use a new dimensionless parameter instead of  $l_\gamma$ . This parameter depends on the power density and is uniquely related to the conversion coefficient.

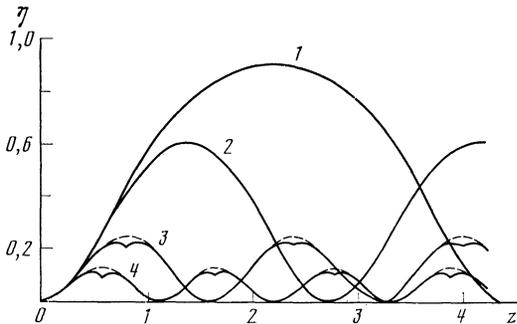


FIG. 1. Comparison of results obtained from (2.3) with the exact solution;  $\Delta s = 0.2, 1.0, 3.0,$  and  $5.0$  for curves 1, 2, 3, and 4, respectively. Broken curves show regions where the SIA differs from the exact solution.

Before we solve (3.1), we must carry out a few derivations. The functions  $A_i$  will be taken in the form

$$\begin{aligned} A_1 &= u_1 \exp\{i\varphi_1(x - \beta_1 z)\}, \quad A_2 = u_2 \exp\{i\varphi_2(x - \beta_2 z)\}, \\ A_3 &= u_3 \exp\{i\varphi_1(x - \beta_3 z) + i\varphi_2(x - \beta_3 z)\}, \end{aligned} \quad (3.2)$$

where  $\varphi_i(x)$  is the phase of the  $i$ th harmonic at entry to the crystal. Thus,  $u_1(0, x)$ ,  $u_2(x, 0)$ ,  $u_3(x, 0)$  are real functions of  $x$  at  $z = 0$ . It is shown in Ref. 13 that the quantities  $|\beta_i(\partial u_i / \partial x)| \sim \beta_i/a$ , which appear on the left-hand side of (3.1) after the substitution of (3.2), can be neglected even for relatively low values of the aperture  $a$ . These quantities will be neglected from now on because, from the practical point of view, we are mostly interested in large-aperture beams ( $\approx 1$  cm). This leads to a set of equations with  $x$ -dependent phase mismatch  $\Delta k(x) = (2\beta_3 - \beta_1 - \beta_2)d\varphi/dx$ . For the boundary conditions  $u_1(0) = u_2(0) = 2^{-1/2}c(x, y, t)$  and  $u_3(0) = 0$ , the conversion coefficient is given by the following expression [analogous to (2.3)]

$$\begin{aligned} \eta(z) &= \frac{1}{Q} \int dx dy dt \left\{ c^2 \text{th}^2(cz) \right. \\ &\quad \left. - \frac{(2\beta_3 - \beta_2 - \beta_1)^2}{32} \left( \frac{d\varphi}{dx} \right)^2 \frac{\text{sh}(zc)}{\text{ch}^3(cz)} \left[ \frac{\text{sh}(4cz)}{4} - cz \right] \right\}, \end{aligned} \quad (3.3)$$

where  $\beta_i = (\beta_i/a)l_{nl}$  are the normalized birefringence angles. It follows from (3.3) that the influence of the dispersion effect on  $\eta$  will vanish altogether when

$$2\beta_3 - \beta_2 - \beta_1 = 0. \quad (3.4)$$

There is a well-known special case of this relation, which arises when the dispersion-theory equations are solved in the first approximation; this is the so-called quasistatic case, for which  $\beta_1 = \beta_2 = \beta_3$  (the group velocities of the interacting waves must satisfy this relation in this case). The most important special case of (3.4) in the theory of harmonic generation in anisotropic media is as follows:

$$\beta_1 = 0, \quad 2\beta_3 - \beta_2 = 0, \quad (3.5)$$

which shows that, when the angle of birefringence at the fundamental frequency is equal to twice the angle of birefringence for the second harmonic, there will be no phase mismatch between the harmonics in interactions of the second type. Conversion will therefore proceed just as effectively as in the case of plane waves.

It is clear from (3.3) that the conversion efficiency is wholly determined by the quantity  $Q^2(x) = (2\beta_3 - \beta_2 - \beta_1)^2 (d\varphi/dx)^2 / 32$ . Let us take the value of  $Q$  at  $x = 2$  so that, for the quadratic phase distribution of the fundamental at entry to the nonlinear medium,

$$A_{1,0}(0, x) = A_0 \exp\left\{ -\left( \frac{x}{a} \right)^2 - i \frac{k}{2R} x^2 \right\} \quad (3.6)$$

we have

$$l = Q(2) = 1/2 [\beta_3 - 1/2(\beta_2 + \beta_1)] k\theta l_{nl}, \quad (3.7)$$

where  $R$  is the phase radius of curvature of the fundamental

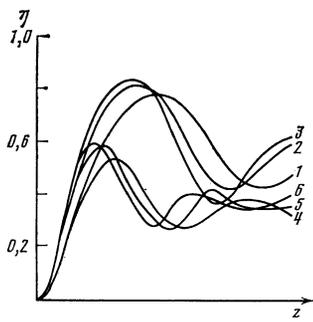


FIG. 2. Conversion coefficient  $\eta$  as a function of  $l = z/l_{ni}$  for beams modulated in space and time;  $l = 0.113$  for curves 1–3 and  $l = 0.513$  for curves 4–6.

at entry into the crystal,  $\theta$  is the measured angle of divergence ( $\theta = 2a/\sqrt{2}R$ ), and  $k$  is the wave number of the first harmonic.

As before, the SIA results were checked against numerical solutions of (3.1). It was found that, for example, for  $l = 0.197$  and  $0.838$ , the  $z$  dependence of  $\eta$  obtained by SIA differed from the numerical result by not more than 1% for values of  $\beta$  for which conversion reached a maximum, and again fell down to almost zero (for example, for  $l = 0.838$ , the conversion coefficient rose from zero to  $0.57$  and then fell to  $0.15$ ).

In contrast to the numerical solution of (3.1) for the two-dimensional system, the SIA method can be used to obtain the conversion coefficient for real beams with an arbitrary distribution of amplitude in all three coordinates. Figure 2 shows the conversion coefficient  $\eta$  for  $l = 0.113$  and  $0.513$  for beams with the following amplitude and phase profile:

$$A_{1,0}(r, 0) = A_0 \exp \left\{ -r^{N_1} - t^{N_2} - i \frac{k}{2R} r^2 \right\}, \quad (3.8)$$

where  $r$  is the position coordinate in the plane perpendicular to the direction of propagation, and  $N_1, N_2$  characterize the respective spatial and temporal components. All the curves in Fig. 2 correspond to  $N_2 = 2$ . Curves 1 and 4 represent Gaussian beams ( $N_1 = 2$ ) and curves 2, 5 and 3, 6 the so-called hypergaussian beam of degree 4 and 6, respectively ( $N_1 = 4$  and 6). It is clear that the curves descend after the maximum (conversion occurs in the reverse direction), and thereafter oscillate, gradually approaching a definite level that is roughly the same for all  $l$ . The oscillations become more pronounced as  $N$  increases. As can be seen, for higher values of  $N_1$ , the conversion maximum appears earlier and is higher. For example, for  $N_1 = 6$ , it is higher by about 6% than for  $N_1 = 2$ . However, this difference declines as  $N_1$  increases.

### 3.1 Rapid estimates of conversion efficiency

As already noted, the single parameter  $l$ , given by (3.7), suffices to enable us to describe the conversion efficiency. In the theory of second-harmonic generation, the question of maximum conversion efficiency ( $\eta_{max}$ ) under given experimental conditions is of considerable practical importance. For beams modulated in both space and time, the relation

between  $\eta_{max}$  and  $l$  can be found by using (2.4) if we replace  $v_b$  with  $\eta_{max}$  and  $\Delta s/4$  with  $l$ :

$$\eta_{max} = (1 + l + 0.5l^2)^{-2}. \quad (3.9)$$

Despite the fact that (2.5) is valid only for plane beams, it is in close agreement with the exact solution for hypergaussian beams with  $N = 6$  and  $N_1 = 2$ . Let us illustrate the practical calculation by the example of the KDP crystal. Two types of wave interaction are possible in this crystal, namely,  $00E$  and  $0EE$ . For the first case,  $\beta_1 = \beta_2 = 0$ ;  $\beta_3 = 2.81 \cdot 10^{-2}$  rad and, for the second,  $\beta_1 = 0$ ,  $\beta_2 = 2.039 \cdot 10^{-2}$  rad,  $\beta_3 = 2.509 \cdot 10^{-2}$  rad. For the same incident power density in the two cases, we have  $l_{ni}(00E)/l_{ni}(0EE) = 1.3$ . Thus, for the same initial conditions, we have the following ratio of the values of  $l$  for the two types of interaction:

$$l_{00E}/l_{0EE} = 2.54. \quad (3.10)$$

Thus, for given power level and given divergence of plane-polarized beams, it is more convenient to perform second-harmonic generation with interaction of the second type provided, of course, the length of the nonlinear medium has been chosen to have the optimum value.

### 3.2. Limiting conversion efficiency for real laser beams

The limiting conversion efficiency that can be achieved in real laser systems is of considerable practical importance, e.g., for the problem of controlled thermonuclear fusion.<sup>14,15</sup> It was shown in Ref. 13 that the angular dispersion effect was the principal factor restricting the conversion efficiency of modern optical frequency-doubling systems. Almost complete (90%) transfer of energy from the fundamental to the harmonic was observed in the experiment with a KDP crystal in Ref. 16. The length of the crystal was 30 mm (angle of birefringence  $\beta = 0.0281$  rad, nonlinearity  $d_{eff} = 1.04 \times 10^{-9}$  esu). The conversion maximum was reached for the power density of  $2.7$  GW/cm<sup>2</sup> and beam aperture of 32 mm. The divergence was about  $6 \times 10^{-5}$  rad. Calculations based on (3.3), (3.7), and (3.9) give good agreement between theory and experiment. Comparison between calculated and measured conversion coefficients shows that the closest fit to the experimental data is achieved for the same parameter values and divergence of  $5.8 \times 10^{-5}$  rad. Calculations show that, for given power density, the limiting (98%) conversion efficiency is achieved only if the divergence is of the order of  $10^{-5}$  rad (if the divergence at exit is set by diffraction), which corresponds to a beam aperture of more than 7 cm. When the limiting conversion efficiency is calculated, it is important to take into account the restricting effect of the depolarization of the fundamental wave. When a real beam is incident on a nonlinear crystal, the polarization at each point in its interior will depart from the optimum direction by an angle  $\psi$ . The maximum conversion coefficient in the case of the  $00E$  interaction will then be

$$\eta_{00E} = \cos^2 \psi \operatorname{th}^2(\cos \psi z) \approx 1 - \psi^2. \quad (3.11)$$

The  $0EE$  conversion coefficient is much more sensitive to a departure from the optimum direction of polarization:

$$\eta_{0EE} = 2 \min(A_0^2, A_e^2) = 2 \sin(\pi/4 - \psi) \approx 1 - 2\psi. \quad (3.12)$$

The resultant conversion coefficient is obtained by taking an average of the individual  $\eta$  over the cross section of the beam:

$$\eta_{max} = \int dx dy dt \eta(\psi, x, y, t). \quad (3.13)$$

Let us define the depolarization coefficient as the ratio of the energy transmitted by the polarizer, whose axis is perpendicular to the optimum direction, to the total energy of the beam. We then have:

$$\varepsilon_{00E} = \varepsilon_{0EE} = \psi^2, \quad \eta_{00E} = 1 - \varepsilon, \quad \eta_{0EE} = 1 - 2\sqrt{\varepsilon}. \quad (3.14)$$

It is clear that, for interactions of the second type, the restriction on the conversion efficiency is stronger than for the first type.

#### 4. NONSTATIONARY INTERACTION OF WAVES

If we use the space-time analogy,<sup>6</sup> we can look upon (3.1) as a set of equations for the triple-frequency interaction of waves in a nonlinear medium with allowance for group delay:

$$\begin{aligned} \frac{\partial A_1}{\partial z} + \frac{1}{u_1} \frac{\partial A_1}{\partial t} &= -i\gamma_1 A_2^* A_3, \\ \frac{\partial A_2}{\partial z} + \frac{1}{u_2} \frac{\partial A_2}{\partial t} &= -i\gamma_2 A_1^* A_3, \\ \frac{\partial A_3}{\partial z} + \frac{1}{u_3} \frac{\partial A_3}{\partial t} &= -i\gamma_3 A_1 A_2, \end{aligned} \quad (4.1)$$

where  $t$  is the time and  $u_i$  are the group velocities of the interacting waves.

As before, we can show that, for the boundary conditions

$$A_1(0) = A_2(0) = A_0/\sqrt{2}, \quad A_3(0) = 0 \quad (4.2)$$

this system reduces to a system with time-dependent phase mismatch:

$$\Delta k(t) = \left( \frac{2}{u_3} - \frac{1}{u_2} - \frac{1}{u_1} \right) \frac{d\varphi}{dt}. \quad (4.3)$$

Let us take the Gaussian pulse with the linear frequency variation

$$A_1(0, t) = A_0 \exp \left[ - \left( \frac{t}{\tau} \right)^2 \left( 1 - \frac{i\Omega_0 \tau}{2} \right) \right] \quad (4.4)$$

as a model of a picosecond pulse. The half-width of this pulse is

$$\Delta\omega = (4 + \Omega_0^2 \tau^2)^{1/2} / \tau. \quad (4.5)$$

By analogy with the foregoing, we then have

$$l = \frac{1}{2\sqrt{2}} \left( \frac{2}{u_3} - \frac{1}{u_2} - \frac{1}{u_1} \right) \Omega_0 l_{nl}. \quad (4.6)$$

For the degenerate interaction ( $\omega_1 = \omega_2 = \omega_3/2$ ), the velocity  $u_3$  in (4.6) must be replaced with the group velocity of the

second harmonic, and we must assume that  $u_1$  and  $u_2$  are both equal to the group velocity of the fundamental.

Since one usually measures the frequency width, it is convenient to express  $l$  in terms of  $\Delta\omega$ . From (4.5), we have

$$l = \frac{1}{2\sqrt{2}} \left( \frac{2}{u_3} - \frac{1}{u_2} - \frac{1}{u_1} \right) l_{nl} \tau^{-1} [(\Delta\omega\tau)^2 - 4]^{1/4}. \quad (4.7)$$

The parameter  $l$  then plays the same rôle as in the effects examined earlier. It has been shown<sup>18</sup> that even a relatively slight frequency deviation will seriously restrict the conversion coefficient for picosecond pulses. Estimates based on calculations of the coherence length  $l_c = (1/u_1 - 1/u_2)^{-1}$  do not then yield satisfactory results.

It was noted in Refs. 6 and 19 that allowance for group delay in the nonlinear equations leads to a substantial increase in the length of the second-harmonic pulse. However, the solution reported in these papers was obtained in the absence of phase mismatch, and the broadening of the pulse was entirely due to the delay between the pulse centers of the two harmonics. On the other hand, it will be shown below that phase modulation can introduce sufficient corrections into this process. The SIA approach leads to the following expression for the shape of the second-harmonic pulse:

$$|A_2|^2 = c^2 \operatorname{th}^2 cz - \frac{l^2 t^2}{4} \frac{\operatorname{sh}(cz)}{\operatorname{ch}^3(cz)} \left[ \frac{\operatorname{sh}(4cz)}{4} - cz \right]. \quad (4.8)$$

The evolution of the Gaussian second-harmonic pulse during its propagation in the nonlinear medium was calculated from (4.8) for  $z = 1, 1.5, 2$ , and  $2.5$ , and the result is shown in Fig. 3. It was assumed that  $l = 1.1$  and that the behavior of  $\eta$  was symmetric with respect to the position of the maximum, as noted in Section 2. The dashed curves show, for comparison, the shape of the second-harmonic pulse for  $l = 0$ . Curve 5 of Fig. 3 corresponds to the hypergaussian pulse ( $N_1 = 6$ ) for  $z = 2.5$ . It is clear from Fig. 3 that the second-harmonic pulse is much narrower (by a factor of 3–4) for  $z > 2$  as compared with the case where phase mismatch was ignored. It is interesting to note that the pulse shape for large  $z$  is practically independent of  $N_1$ , i.e., it is not sensitive to the shape of the incident radiation.

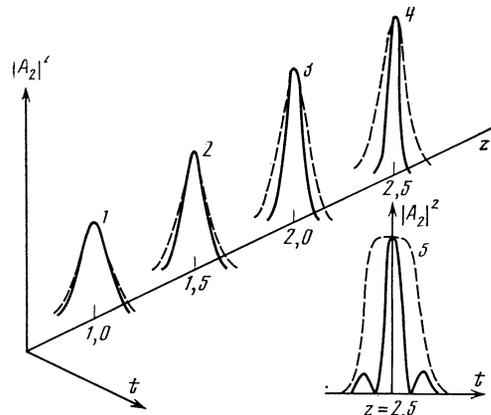


FIG. 3. Evolution of a pulse during its propagation in a nonlinear medium. Curves 1–4 correspond to the Gaussian pulse ( $N_2 = 2$ ) and curve 5 to the hypergaussian pulse ( $N_2 = 6$ ).

It is important to note that the curves in Fig. 3 were plotted without allowing for the spreading of the pulse due to group delay. In general, there will be two competing processes, one of which stretches and the other compresses the harmonic pulse. Thus, for large values of the dimensionless parameter  $l$  and coherence length  $l_c$ , the pulse compression will predominate, whereas, in the opposite case, pulse stretching will do so.

## 5. EFFECT OF CUBIC NONLINEARITY ON THE INTERACTION PROCESS IN QUADRATIC MEDIA

The power density being used in the nonlinear processes has shown an upward trend. This means that higher-order nonlinearities (above all, the cubic nonlinearity) may become superimposed on the quadratic interaction. The truncated equations describing interactions in a quadratic medium with nonzero cubic susceptibility then assume the form

$$\begin{aligned} \frac{dA_1}{dz} &= -i\gamma A_1^* A_2 e^{-i\Delta s z} - i\theta_1 |A_1|^2 A_1 - i2\theta_2 |A_2|^2 A_1, \\ \frac{dA_2}{dz} &= -i\gamma A_1^2 e^{i\Delta s z} - i4\theta_2 |A_1|^2 A_2 - i2\theta_3 |A_2|^2 A_2, \end{aligned} \quad (5.1)$$

where  $\theta_i$  are the third-order nonlinearity coefficients. Simple substitution will reduce (5.1) to the following form:

$$\frac{du}{dz} = -iu^*v, \quad \frac{dv}{dz} = -iv^2 - i\{\Delta s_0 + 2(\beta_1 - \beta_2 |u|^2)\}v, \quad (5.2)$$

where  $A_0$  is the amplitude of the fundamental at entry to the nonlinear medium,

$$\Delta s_0 = \Delta k l_{nl}, \quad \beta_1 = (\theta_3 - 2\theta_2) |A_0|^2 l_{nl}, \quad (5.3)$$

$$\beta_2 = (\theta_1 + \theta_3 - 4\theta_2) |A_0|^2 l_{nl},$$

and  $z$  is normalized to  $l_{nl}$ .

Solving (5.2) in the strong-interaction approximation, in accordance with (1.4) and (1.5), we obtain

$$\begin{aligned} \eta(z) &= \text{th}^2 z - \varphi_1 \frac{\text{sh} z}{\text{ch}^3 z} \left[ \frac{\text{sh} 4z}{4} - z \right] \\ &\quad - \varphi_2 \frac{\text{sh}^4 z}{\text{ch}^2 z} + \varphi_3 \text{th}^4 z \text{ch} 2z, \end{aligned} \quad (5.4)$$

where

$$\begin{aligned} \varphi_1 &= \frac{1}{8} \left\{ \left[ \beta_1 + \frac{\Delta s_0 + \beta_2}{2} \right]^2 - 6\beta_2 \left( \beta_1 + \frac{\Delta s_0}{2} \right) \right\}, \\ \varphi_2 &= \beta_2 \left( \beta_1 + \frac{\Delta s_0 + \beta_2}{2} \right), \quad \varphi_3 = \frac{\beta_2^2}{4}. \end{aligned}$$

The optimization of the process is, of course, an interesting question. If we allow  $z$  in (5.4) to tend to infinity, the condition  $\eta \rightarrow 1$  will yield

$$\Delta s_{\text{opt}} = \beta_2 - 2\beta_1, \quad (5.5)$$

where  $\Delta s_{\text{opt}}$  is the optimum phase mismatch that completely compensates the cubic nonlinearity. The expression given by (5.5) was checked by a numerical calculation and was fully confirmed.

It is important to note that, if  $\Delta s_{\text{opt}}$  is determined from (5.5), 100% conversion is possible only for small  $\beta$ , i.e., in the region in which SIA is still valid. Numerical calculations show that (5.5) will no longer ensure complete conversion when  $\beta_1 = \beta_2/z < 1$ . It must also be remembered that the foregoing applies only to planar beams with uniform amplitude distribution. When the inhomogeneity in the amplitude distribution of the radiation incident on the nonlinear medium is taken into account, the compensation of the effect of the cubic nonlinearity by phase mismatching will not be complete. The value of  $\Delta s_{\text{opt}}$  for modulated beams will be somewhat different from that for planar beams. When the beam shape is given by (3.8), the biggest difference will occur for Gaussian beams:  $N_1 = N_2 = 2$ . As  $N_1, N_2$  increase, the optimum phase mismatch for spatially, inhomogeneous beams will approach the optimum value for planar beams. There will also be a corresponding increase in the gain in efficiency for large  $N_1, N_2$  as compared with small values of these quantities. Each point in Fig. 4 corresponds to the maximum conversion coefficient  $\eta_{\text{max}}$  that can be reached for given  $\Delta s_0$ ,  $\beta_1, \beta_2$ . Curves 1, 2, and 3 are plotted for  $\beta_1 = 0.1$ ,  $\beta_2 = 0.4$ , and curves 4, 5, and 6 for  $\beta_1 = 0$ ,  $\beta_2 = 0.4$ . The horizontal axis shows points at which the maximum in  $\Delta s_0$  is reached in the case of plane waves. As can be seen, the optimum  $\Delta s_0$  for modulated beams is not very different from that for unmodulated beams, but the height of the maximum is considerably lower.

## 6. INTERACTION OF WAVES IN LINEARLY INHOMOGENEOUS MEDIA

The course of nonlinear processes depends in many respects on the quality of the crystals used, since crystals frequently exhibit considerable growth inhomogeneity in their birefringence in the longitudinal direction. Let us consider stationary triple-frequency interaction in a weakly inhomogeneous medium in which the refractive index is a linear function of  $z$  (this approximation is close to the practical situation<sup>20</sup>):

$$\frac{dA_1}{dz} = -i\gamma A_1^* A_2, \quad \frac{dA_2}{dz} = i\Delta k_0 A_2 - i\Delta k_1 z A_2 = -i\gamma A_1^2. \quad (6.1)$$

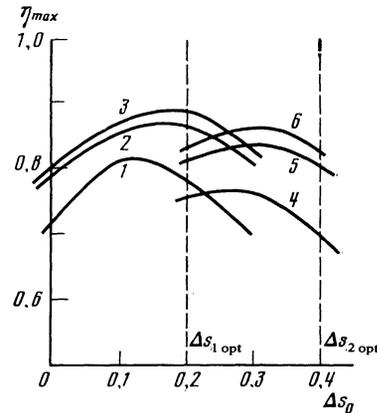


FIG. 4. Maximum conversion coefficient as a function of  $\Delta s_0$ : 1, 4—Gaussian beams; 2, 5—hypergaussian beams ( $N_1 = 4$ ,  $N_2 = 2$ ); 3, 6—hypergaussian beams ( $N_1 = 6$ ,  $N_2 = 2$ );  $\Delta s_{1 \text{ opt}}$ ,  $\Delta s_{2 \text{ opt}}$ —optimum values of  $\Delta s_0$  for unmodulated waves.

Here  $\Delta k_0$  and  $\Delta k_1$  denote the constant and variable phase mismatch, respectively. SIA then gives the following expression for the conversion coefficient:

$$\eta(z) = \text{th}^2 z - \frac{(\Delta \tilde{k}_0)^2}{32} \frac{\text{sh } z}{\text{ch}^3 z} \left[ \frac{\text{sh}(4z)}{4} - z \right] + \frac{\Delta \tilde{k}_0 \Delta \tilde{k}_1}{4} F_1(z) - \frac{(\Delta \tilde{k}_1)^2}{4} F_2(z),$$

$$F_1(z) = z^2 \frac{\text{sh } z}{\text{ch}^3 z} + 2z \text{th}^2 z - 2 \text{th}^3 z - \frac{\text{sh}^3 z}{\text{ch}^3 z},$$

$$F_2(z) = \frac{4}{3} z^3 \frac{\text{sh } z}{\text{ch}^3 z} - 4 \frac{z^2}{\text{ch}^2 z} + \frac{7}{2} z \frac{\text{sh } z}{\text{ch}^3 z} - 4z \text{th } z + \text{th}^2 z \left[ \frac{\text{ch}(2z)}{2} + 4 \right], \quad (6.2)$$

where  $\Delta \tilde{k}_0 = \Delta k_0 l_{\text{nl}}$  and  $\Delta \tilde{k}_1 = \Delta k_1 l_{\text{nl}}^2$ . It is clear that, as the power density increases, the contributions of the second and third terms in (6.2) will decrease as compared with the first. Consequently, the effect of the inhomogeneity of the medium on the course of processes occurring in it can be eliminated by increasing the incident power density. As before, let us consider the function (6.2) for large  $z$ . Setting to zero the sum of all terms proportional to  $\Delta k_1$ , we obtain

$$\Delta \tilde{k}_0 = -2\Delta \tilde{k}_1 \quad (6.3)$$

and this relation between the constant and variable phase-mismatch coefficients will ensure that the conversion efficiency will tend to 100%. The validity of these conclusions has been confirmed by numerical calculations for a broad range of values of  $\Delta \tilde{k}_0$  and  $\Delta \tilde{k}_1$ .

Expression (6.3) becomes exact when the spatial inhomogeneity in the amplitude of a real beam is taken into account. Actually, (6.3) will not immediately apply to all the rays at once because the normalized coefficients  $\Delta \tilde{k}_0$  and  $\Delta \tilde{k}_1$  depend on  $l_{\text{nl}}$ . However, despite the considerable restriction imposed on conversion, the optimum phase mismatch will not be very different from the value given by (6.3). Thus, instead of  $\Delta \tilde{k}_0 = -0.8$ , given by (6.3) for  $\Delta \tilde{k}_1 = 0.4$ , we obtain  $\Delta \tilde{k}_0 = -0.9$ . As for the effect of the beam shape on conversion, we have the same result as before: because of the considerable inhomogeneity of hypergaussian as compared with Gaussian beams, the conversion efficiency will be high in this case as well.

## CONCLUSION

We have developed a theory of nonlinear waves in the strong-interaction approximation. Application of this approximation to nonlinear processes involving intensive energy transfer between real waves in real media is physically more valid than the use of the other approximations employed so far, especially since almost complete transfer of pump energy to the harmonic has now been produced experimentally.<sup>16</sup>

The potentialities of the method that we have developed were demonstrated above by considering the example of sec-

ond-harmonic generation by real wave beams, and by pulses in homogeneous and inhomogeneous media. Numerical solution of the nonlinear equations on a computer has shown that SIA results in high precision not only in the case of a strong interaction, but also when it is highly restricted. Analysis of the frequency-multiplication process in the case of wave beams in a dispersive medium has enabled us to derive an expression for the dimensionless parameter characterizing the influence of the dispersion effect. Estimates using the parameter  $l$  show that conversion can be highly restricted even in those regions where quantities previously used in estimates do not lead to any restriction on conversion. A simple formula involving  $l$  has been obtained and can be used to predict to within a few percent the maximum conversion coefficient that can be achieved experimentally for a beam modulated in the three coordinates. The most important result of SIA is the simple expression for the conversion efficiency that is convenient in calculations and can be used in rapid estimates of which particular interaction type predominates in a given case. Our estimate of the effect of the depolarization factor on wave conversion efficiency in KDP has shown that the restriction imposed on the process is greater for the *OEE* than the *OOE* interaction.

For higher power densities, the nonlinear interaction in a quadratic medium may be restricted by cubic nonlinearity. Analysis of this process has shown that this restriction can be compensated by suitably choosing the optimum phase mismatch.

In the case of nonstationary frequency multiplication and mixing, conversion efficiency can saturate for near-zero group velocity difference and slight phase modulation of the ultrashort pulse. We note that the harmonic pulse can then be much shorter than the pump pulse.

The potentialities of SIA are not exhausted by the application examined above. The approximation is, in principle, suitable for a broad range of problems in the theory of nonlinear waves when parameters analogous to  $\beta$  are small.

<sup>1</sup>S. A. Akhmanov and R. V. Khokhlov, *Problemy nelineinoi optiki* (Problems in Nonlinear Optics), Academy of Sciences of the USSR, 1964.

<sup>2</sup>V. N. Tsytovich, *Nelineinnye efekty v plazme* (Nonlinear Effects in Plasmas), Nauka, Moscow, 1967.

<sup>3</sup>O. V. Rudenko and S. N. Soluyan, *Teoreticheskie osnovy nelineinoi akustiki* (Theoretical Foundations of Nonlinear Acoustics), Nauka, Moscow, 1975.

<sup>4</sup>N. Bloembergen, *Nonlinear Optics*, Benjamin, 1965 [Russian translation, Mir, Moscow, 1966].

<sup>5</sup>S. A. Akhmanov, A. P. Sukhorukov, and A. S. Chirkin, *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **10**, 1639 (1967).

<sup>6</sup>S. A. Akhmanov and A. S. Chirkin, *Statisticheskie yavleniya v nelineinoi optike* (Statistical Phenomena in Nonlinear Optics), Moscow State University, 1971, p. 25.

<sup>7</sup>Z. A. Tagiev and A. S. Chirkin, *Zh. Eksp. Teor. Fiz.* **73**, 1271 (1977) [Sov. Phys. JETP **46**, 669 (1977)].

<sup>8</sup>Z. A. Tagiev and A. S. Chirkin, *Kvantovaya Elektron. (Moscow)* **7**, 1337 (1980) [Sov. J. Quantum Electron. **10**, 766 (1980)].

<sup>9</sup>V. G. Dmitriev and L. V. Tarasov, *Prikladnaya nelineinaya optika* (Applied Nonlinear Optics), *Radio i Svyaz*, 1982, p. 123.

<sup>10</sup>Yu. N. Karamzin and A. P. Sukhorukov, *Zh. Eksp. Teor. Fiz.* **68**, 834 (1975) [Sov. Phys. JETP **41**, 414 (1975)].

- <sup>11</sup>L. P. Mel'nik, I. N. Filonenko, and A. I. Kholodnykh, *Kvantovaya Elektron. (Moscow)* **6**, 25 (1979) [*Sov. J. Quantum Electron.* **9**, 13 (1979)].
- <sup>12</sup>A. I. Filatov and L. V. Sharova, *Integral'nye neravenstva i teoriya nelineinykh kolebaniy (Integral Inequalities and the Theory of Nonlinear Oscillations)*, Nauka, Moscow, 1976.
- <sup>13</sup>E. A. Ibragimov, V. I. Redkorechev, A. P. Sukhorukov, and T. Usmanov, *Kvantovaya Elektron. (Moscow)* **9**, 1131 (1982) [*Sov. J. Quantum Electron.* **12**, 714 (1982)].
- <sup>14</sup>F. Amiranoff, R. Benattar, and R. Farbo, *Bull. Am. Phys. Soc.* **24**, 1069 (1979).
- <sup>15</sup>S. E. Max and R. Estabrook, *Comments Plasma Phys. Fusion* **5**, 239 (1980).
- <sup>16</sup>A. A. Gulamov, E. A. Ibragimov, V. I. Redkorechev, and T. Usmanov, *Kvantovaya Elektron. (Moscow)* **10**, 1305 (1983) [*Sov. J. Quantum Electron.* **13**, 844 (1983)].
- <sup>17</sup>M. B. Vinogradova, O. V. Rudenko, and A. P. Sukhorukov, *Teoriya voln (Theory of Waves)*, Nauka, Moscow, 1979, p. 94.
- <sup>18</sup>E. A. Ibragimov and T. Usmanov, *Dokl. Akad. Nauk SSSR* **261**, 846 (1981) [*Sov. Phys. Dokl.* **26**, 1138 (1981)].
- <sup>19</sup>G. V. Krivoshechekov, I. G. Nikulin, and R. I. Sokolovskii, *Opt. Spektrosk.* **31**, 448 (1971) [*Opt. Spectrosc. (USSR)* **31**, 238 (1971)].
- <sup>20</sup>F. R. Nach, G. D. Boyd, M. Sargent III, and P. M. Bridenbaugh, *J. Appl. Phys.* **41**, 2564 (1970).

Translated by S. Chomet