THEORY OF GENERATION OF OPTICAL HARMONICS IN CONVERGING BEAMS

S. A. AKHMANOV, A. P. SUKHORUKOV, and R. V. KHOKHLOV

Moscow State University

Submitted to JETP editor August 25, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) 50, 474-486 (February, 1966)

A consistent theory of nonlinear optical effects in bounded beams is developed. The theory is based on the parabolic equation method extended to nonlinear problems. This approach can be employed for analyzing nonlinear wave processes by taking into account diffraction effects in first-order approximation. The generation of second optical harmonics by a weakly converging cylindrical wave in an anisotropic medium is considered as an example. The intensity and spatial structure of the harmonics are determined. Satisfactory agreement between the theory and experiment is noted.

1. INTRODUCTION

I. IN spite of the considerable practical interest that attaches to nonlinear optical effects, a more or less satisfactory theory of interacting electromagnetic waves in a nonlinear medium has been developed only for plane waves (one-dimensional problems, unbounded beams). Although its results can be used in some cases (by introducing a few correction factors) to interpret nonlinear-optics experiments on real bounded beams from lasers, there are many experiments for which a simplified approach is impossible, especially with focused beams. An exact analysis of nonlinear effects at the focus (for example, the generation of optical harmonics, induced scattering) makes it necessary to take into account at least the twodimensional character of the interacting beams. It must also be emphasized that only a consistent theory of nonlinear interaction of bounded beams makes it possible to formulate quantitative criteria for the applicability of the theory of plane waves to an analysis of phenomena in unfocused laser beams.

2. Experiments on the generation of optical harmonics in focused beams were carried out during the early development of nonlinear optics (see, for example, ^[11]). The first attempt to develop a theory for the generation of the second optic harmonic in a focus was undertaken by Kleinman, ^[2] but his approach was excessively oversimplified. Kleinman confined himself essentially to consideration of only the region directly adjacent to the focal plane, where the wave can be regarded as plane and the nonlinear interactions one-dimensional. Experiments described in ^[3] have yielded

data essentially different from Kleinman's calculations; the greatest differences appeared in the distribution of the intensity harmonic over the beam cross section. It is shown in ^[3] that an account of one-dimensional nonlinear interactions alone is insufficient for a correct interpretation of the experimental data and that allowance must be made for two-dimensional interactions.

Thus, for a correct description of nonlinear effects at the focus it is obviously necessary to consider the whole problem, that is, to analyze the character of development of the nonlinear process in the entire region occupied by the beam, and not only in the direct vicinity of the focal plane.

3. The propagation of electromagnetic waves in a nonlinear medium is described by an equation of the form

$$[\nabla [\nabla \mathbf{E}]] + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}^{(l)}}{\partial t^2} + \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}^{(nl)}}{\partial t^2} = 0, \quad (1)^*$$

where

$$\mathbf{P}^{(l)} = \int_{0}^{\infty} \hat{\boldsymbol{x}}(t') \mathbf{E}(t-t') dt', \qquad (2)$$

$$\mathbf{P}^{(\mathbf{n}l)} = \int_{0}^{\infty} dt' \int_{0}^{\infty} dt'' \,\chi(t',t'') \,\mathbf{E}(t-t') \,\mathbf{E}(t-t'-t'') + \dots,$$
(3)

 κ and $\hat{\chi}$ are tensors of second, third, and higher rank. For the case of plane waves (for simplicity unmodulated) the procedure for analyzing the nonlinear equation (1) consists of a simplification that reduces to a neglect of the second derivatives with respect to the coordinates (the amplitudes and the

*[
$$\nabla \mathbf{E}$$
] = $\nabla \times \mathbf{E}$.

phases of the interacting waves are assumed to be slowly varying functions of the coordinates—see, for example, ^[4]). The solution of (1) is sought in the form

$$\mathbf{E} = \sum_{n} \mathbf{e}_{n} A_{n}(\mu \mathbf{r}) \exp\{i(\omega_{n} t - \mathbf{k}_{n} \mathbf{r})\}, \qquad (4)$$

where μ is a small parameter with magnitude $\sim A/A_{at}$, A_{at} being the atomic field. Substituting (4) into (1) and retaining only terms proportional to $\sim \mu$ (the nonlinear and dissipative terms in (1) are of this order), we arrive at the system of so-called abbreviated equations, which describe interaction of plane waves in a nonlinear medium:

$$\left[\mathbf{e}_{n}\left[\mathbf{k}_{n}\mathbf{e}_{n}\right]\right]\nabla A_{n}+\mathbf{e}_{n}\hat{\boldsymbol{\alpha}}\mathbf{e}_{n}A_{n}=F_{n^{(2)}}(A_{l},A_{m})+\ldots,\quad(5)$$

where $\hat{\alpha}$ is the conductivity tensor of the medium and $F_n^{(k)}$ are terms connected with the nonlinear polarization. Most theoretical papers on nonlinear optics published to date are based on equations such as (5). These equations characterize, as can be readily seen, the variation of the amplitudes of the interacting waves along the rays—the vector $[\mathbf{e_n} \times [\mathbf{k_n} \times \mathbf{e_n}]]$ is parallel to the ray vector $\mathbf{s_n}$. Thus the use of equations such as (5) for an analysis of phenomena in bounded beams (this approximate approach is the basis of papers ^[2-4]) corresponds essentially to the geometric-optics approximation.

To allow for diffraction effects in a beam of finite cross section, it is necessary to introduce the derivatives of the amplitude not only along the rays, but also transverse to them. Since a transition to the shadow region takes place in directions perpendicular to the beam, the variations of the amplitude along the beam should be regarded as slower than those transverse to the beam. It is therefore natural to seek for a bounded beam a solution of (1) in the form

$$\mathbf{E} = \sum_{n} \mathbf{e}_{n} A_{n} (\mu \mathbf{r} \mathbf{s}_{n}, \, \overline{\mathbf{\gamma} \mu} \, [\mathbf{r} \mathbf{s}_{n}]) \exp\{i(\omega_{n} t - \mathbf{k}_{n} \mathbf{r})\}.$$
(6)

Substituting (6) in (1) we obtain in first approximation in μ , in place of (5), the partial differential equation

$$[\mathbf{e}_n[\mathbf{k}_n\mathbf{e}_n]]\nabla A_n + \frac{\iota}{2}\Delta_{\perp}A_n + \mathbf{e}_n\hat{\alpha}\mathbf{e}_n = F_n^{(2)}(A_l, A_m) + \dots,$$
(7)

where Δ_{\perp} is the Laplacian corresponding to differentiation in a direction perpendicular to the beam.

In a linear medium (all $F_n^{(k)} = 0$) Eq. (7) goes over into an equation of the parabolic type with an imaginary "diffusion" coefficient; such an equation is extensively used in diffraction theory. By now the parabolic-equation method has been used in many problems involving diffraction in linear isotropic^[5-7] and anisotropic media.^[8] We shall use equations of the (7) type and analyze in detail the generation of the second harmonic of a cylindrical wave in a medium whose polarization depends quadratically on the field intensity.

2. REPRESENTATION OF A WEAKLY CONVERGING CYLINDRICAL WAVE

We shall represent the general formula for an unmodulated wave that converges cylindrically to a point (see, for example, ^[9, 10])

$$\mathbf{E}(x, z, t) = \int_{-\infty}^{+\infty} \mathbf{f}(a) \exp\{i\left[\omega t - kr\cos(\theta - a)\right]\} da \qquad (8)$$

(where r and θ are the polar coordinates in the x,z plane), for the case of a weakly converging wave (the function $\mathbf{f}(\alpha)$ differs from zero for angles $|\alpha| < \alpha' \sim \sqrt{\mu}$), in the form

1

$$\mathbf{E} = \mathbf{e}A\left(x, z\right) \exp\left\{i\left(\omega t - kz\right)\right\},\tag{9a}$$

$$A(x, z) = \sqrt{\frac{P_{\mathbf{i}}k}{cba'}} \int_{-\alpha'}^{\alpha'} \exp\left\{-ikxa + \frac{ika^2}{2}z\right\} da,$$
$$P_{\mathbf{i}} = \frac{cb}{4\pi} \int_{-\infty}^{\infty} AA^{\bullet} dx, \qquad (9b)$$

where P_1 is the total power of the beam and b the width of the beam in the direction perpendicular to the x,z plane; when $b >> \lambda$ the diffraction can be disregarded in this direction.

The representation (9) can be regarded as a satisfactory model of a laser beam focused with the aid of a relatively long-focus cylindrical lens, and also an unfocused beam generated by a flat crystal. It is also easy to verify that (9) corresponds fully to the approximations used in a diffraction theory based on an analysis of the parabolic equation; the slowly varying amplitude (9b) satisfies a linear equation such as (7).

The amplitude of a cylindrically converging wave reaches a maximum on a focal line of length b (x = 0, z = 0) and is equal to

$$A_{\rm f} = 2\gamma \overline{P_1 k \alpha' / cb}; \qquad (10)$$

in the x,z plane the focal spot is in the form of an ellipse with semi-axes

$$z_{\rm f} = \lambda / (\alpha')^2, \quad x_{\rm f} = \lambda / 2\alpha'.$$
 (11)

The variation of the structure of the field of the focused beam was analyzed in detail in [9, 10]. In the direct vicinity of the focal plane z = 0, the wave can be regarded with sufficient accuracy as plane; on the boundary of the focal spot, and with

increasing distance from it, the front becomes more and more cylindrical.

3. DERIVATION OF PARABOLIC EQUATIONS DESCRIBING THE EXCITATION OF A SECOND HARMONIC BY CYLINDRICAL WAVES IN AN ANISOTROPIC MEDIUM

With the aim of comparing the results of the theory with experiment, let us consider the problem of greatest practical interest, that of generation of a second harmonic in a uniaxial negative crystal, by means of an ordinary wave of fundamental radiation (see, for example, the experimental papers ^[1, 3, 11]) whose frequency is far from any absorption line. However, unlike in ^[2, 4], we assume that the fundamental wave is not plane but has the form (9). The harmonic-generation process occurs most effectively near the so-called direction of one-dimensional synchronism, along which the phase velocities of the ordinary wave of the fundamental radiation and of the extraordinary wave of the second harmonic coincide. Then, in accordance with (6) and (9), the field in the nonlinear medium can be represented in the form

$$\mathbf{E} = \mathbf{e}_{1}^{0}A_{1}(\mu z_{0}; \, \forall \mu x_{0}) \, \exp \left\{ i \left(\omega t - k_{1}^{0} z_{0} \right) \right\} \\ + \, \mathbf{e}_{2}^{e}A_{2}\left(\mu z_{e}; \, \forall \overline{\mu} x_{e} \right) \, \exp \left\{ i \left(2\omega t - k_{2}^{e} z_{e} \right) \right\} + \, \mathbf{c. c.}$$
(12)

In the nonlinear medium there interact a cylindrical ordinary wave of fundamental frequency (polarization vector \mathbf{e}_1^0 , wave vector \mathbf{k}_1^0 , ray vector \mathbf{s}_1^0 , axis \mathbf{z}_0 parallel to \mathbf{s}_1^0 , $\mathbf{x}_0 \parallel \mathbf{e}_1^0$) and a cylindrical extraordinary wave of the second harmonic ($\mathbf{e}_2^e, \mathbf{k}_2^e, \mathbf{s}_2^e; \mathbf{z}_e \parallel \mathbf{s}_2^e; \mathbf{x}_e \parallel \mathbf{e}_2^e$). Assuming that the axis of the fundamental beam coincides with the direction of one-dimensional synchronism (along which $\mathbf{k}_2^e = 2\mathbf{k}_1^0$), substituting (12) in (1), retaining only the first term of (3), assuming $\hat{\kappa}$ to be a real quantity, and retaining terms $\sim \mu$, we obtain for the complex amplitudes A_1 and A_2 two equations of the parabolic type in terms of the corresponding ray coordinate:

$$\frac{\partial A_1}{\partial z_0} = \frac{1}{2ik_{1^0}} \frac{\partial^2 A_1}{\partial x_{0^2}} + \frac{4\pi\omega^2}{ic^2 k_{1^0}} (\mathbf{e_1}^0 \hat{\boldsymbol{\chi}}_{2^{\omega-\omega}} \mathbf{e_2}^e \mathbf{e_1}^o) A_2 A_1^*, \quad (13a)$$
$$\frac{\partial A_2}{\partial z_e} = \frac{1}{2ik_{2^e}} \frac{\partial^2 A_2}{\partial x_{e^2}} + \frac{8\pi\omega^2}{ic^2 k_{2^e} \cos \mathbf{k_2}^e \mathbf{s_2}^e} (\mathbf{e_2}^e \hat{\boldsymbol{\chi}}_{2^{\omega}} \mathbf{e_1}^0 \mathbf{e_1}^o) A_1^2. \quad (13b)$$

Here $\hat{\chi}^{2\omega}$ and $\hat{\chi}^{2\omega-\omega}$ are the spectral components of the tensor $\hat{\chi}$. Equations (13) are simpler than the initial equation (1), but even their solution entails great difficulty in the general case. (We note incidentally that (13) are more convenient than (1) for digital-computer solution.)

To obtain analytic results, we simplify (2) fur-

ther, confining ourselves to the given-field approximation (we neglect the reaction of the second harmonic on the fundamental radiation), and write both equations of (13) in terms of the ray coordinates x_0 and z_0 . The latter can be easily done by intro-

ducing the anisotropy angle $\beta = \widehat{\mathbf{k}_2^e \mathbf{s}_2^e}$; in accordance with the foregoing, $\widehat{\mathbf{s}_1^0 \mathbf{s}_2^e} = \widehat{\mathbf{k}_2^e \mathbf{s}_2^e} = \beta$ and consequently

$$z_0 = z_e \cos \beta - x_e \sin \beta, \quad x_0 = z_e \sin \beta + x_e \cos \beta.$$
 (14)

Carrying out the transformations and recognizing that the anisotropy angle can be regarded as a small quantity $\beta \sim \sqrt{\mu}$, we arrive at the equations

$$\frac{\partial A_1}{\partial z_0} = \frac{1}{2ik_1^0} \frac{\partial^2 A_1}{\partial x_0^2},$$
 (15a)

$$\frac{\partial A_2}{\partial z_0} = \frac{1}{2ik_2^e} \frac{\partial^2 A_2}{\partial x_0^2} - \beta \frac{\partial A_2}{\partial x_0} - i\gamma A_1^2, \qquad (15b)$$

where

$$\gamma = \frac{8\pi\omega^2}{c^2k_2^e} (\mathbf{e}_2^e \hat{\boldsymbol{\chi}}^{2\omega} \, \mathbf{e}_1^{0} \mathbf{e}_1^{0}).$$

Equations such as (15) correspond to a definite approximation in the calculation of the dispersion properties of the nonlinear medium. Within the framework of (15), the equation for the sections of the wave vectors can be obtained (compare with ^[81]) by putting $\gamma = 0$ and

$$A_{1} = \exp \left\{-i(q_{x}^{0}x_{0} + q_{z}^{0}z_{0})\right\},$$
(16a)

$$A_2 = \exp \{-i(q_x^e x_0 + q_z^e z_0)\}.$$
 (16b)

Substituting (16a) in (15a) we obtain for the ordinary wave the equation of a parabola

$$q_z^0 + \frac{1}{2k_1^0} (q_x^0)^2 = 0, \qquad (17a)$$

and for the extraordinary wave (substituting (16b) in (15b))

$$q_z^e + \frac{1}{2k_2^e}(q_x^e)^2 + \beta q_x^e = 0.$$
 (17b)

The parabolas (17) approximate the sections of the real surfaces of the wave vectors—circle and ellipse (see Fig. 1).

4. GENERAL SOLUTION OF THE PARABOLIC EQUATION OF THE SECOND HARMONIC FOR A SPECIFIED FUNDAMENTAL-RADIATION FIELD. CONDITIONS OF EFFECTIVE GENERATION OF THE HARMONIC

Assume that a cylindrically converging wave of fundamental radiation, defined by (9), is incident



FIG. 1. Sections of the surfaces of the wave numbers of the ordinary wave of fundamental frequency (curve 1°) and extraordinary wave of the second harmonic (curve 2°). Synchronous one-dimensional interactions occur along the z axis; we also show here a diagram of the synchronous two-dimensional interaction.

on a layer of nonlinear medium of thickness L (Fig. 2) in such a way that the focus is located at a distance l_1 and l_2 from the front and rear boundaries of the layer, respectively ($l_1 + l_2 = L$). We align the x and z axes with x_0 and z_0 , and locate the origin at the focus. We assume that $A_2 = 0$ when $z = -l_1$ and the amplitude of the refracted wave of fundamental frequency in the medium (which is a solution of (15a)) is

$$A_{1}(x,z) = \sqrt{\frac{P_{1}k_{1}}{cba_{0}}} \int_{-\alpha_{0}}^{\alpha_{0}} \exp\left\{-ik_{1}^{0}x\alpha + \frac{ik_{1}^{0}\alpha^{2}}{2}z\right\} d\alpha. \quad (18)$$

In (18) the angles α are measured from the beam axis in the nonlinear medium; the integration in (18) is within the limits $[-\alpha_0; \alpha_0]$ —the angular aperture of the beam in the medium, $\alpha_0 = \alpha' k_0 / k_1^0$, where k_0 is the wave number for the fundamental wave in the linear medium (usually

 $k_0/k_1^0 < 1$ and $\alpha_0 < \alpha'$, see Fig. 2). Under the formulated boundary conditions, the amplitude of the second harmonic on the rear face of the nonlinear crystal ($z = l_2$) is

$$A_{2}(x, l_{2}) = -i\gamma \int_{-l_{1}}^{l_{2}} d\tau \sqrt{\frac{ik_{2}^{e}}{2\pi(l_{2}-\tau)}} \int_{-\infty}^{\infty} d\xi A_{1}^{2}(\xi, \tau)$$
(19)

$$\times \exp\left\{\frac{-ik_{2}^{e}[\xi - x + \beta(l_{2}-\tau)]^{2}}{2(l_{2}-\tau)}\right\}$$

Substituting here A_1 from (18) and integrating with respect to ξ , we obtain

$$A_2(x, l_2) = \frac{-i\gamma P_1 k_1^0}{cba_0} \exp\left\{\frac{-ik_2^2 l_2 \beta^2}{2}\right\} \int_{-l_1}^{l_2} d\tau \int_{-\alpha_0}^{\alpha_0} d\alpha_1 \int_{-\alpha_0}^{\alpha_0} d\alpha_2$$



FIG. 2. Diagram explaining second-harmonic generation by a cylindrically converging lens in a layer of a nonlinear medium of length $l_1 + l_2 = L$. The normal to the boundary of the layer coincides with the beam axis and with the synchronous direction for the one-dimensional interaction; α' is the divergence of the beam in the linear isotropic medium, $\alpha_0 = \alpha' k_0/k_1^0$ is the divergence in the nonlinear medium; R is the shift of the focus in the nonlinear medium.

$$\times \exp\left\{-ik_{1}^{0}x(\alpha_{1}+\alpha_{2})+\frac{il_{2}(k_{2}^{e}\beta+k_{1}^{0}\alpha_{1}+k_{1}^{0}\alpha_{2})^{2}}{2k_{2}^{e}}+\frac{ik_{1}^{0}\sigma}{4}[(\alpha_{1}-\alpha_{2})^{2}-4\beta(\alpha_{1}+\alpha_{2})]\right\}.$$
(20)

The behavior of the second harmonic at a point of observation behind the rear face of the nonlinear medium is described when $z > l_2$ by a parabolic equation of the form (15a), in which k_1^0 should be replaced by $2k_0$ —the wave number of the second harmonic in the linear medium, which in turn is assumed isotropic (for example, vacuum). The solution of the Cauchy problem for an equation of the type (15a) can be written either by using expression (2) directly, or by using the angular spectrum of the second-harmonic amplitude in the section l_2 , i.e., a representation of the type (9). The latter (see, for example, ^[71]; cf. also formula (9)) is of the form

$$\begin{aligned} A_2(x,z) &= \int_{-\infty}^{\infty} A_2(k_{2x}, l_2) \\ &\times \exp\left\{-ik_{2x}x + \frac{ik_{2x}^2(z-l_2)}{4k_0}\right\} dk_{2x} \quad (z \ge l_2), \quad (21) \end{aligned}$$

where

2

$$A_2(k_{2x}, l_2) = \frac{1}{2\pi} \int_{-\infty}^{\infty} A_2(x, l_2) \exp\{ik_{2x}x\} dx.$$
 (22)

By applying the transformation (22) to (20) we obtain

$$A_{2}(k_{2x}, l_{2}) = \frac{-i\gamma P_{1}k_{1}^{0}}{cba_{0}} \exp\left\{\frac{-ik_{2}e^{l}_{2}\beta^{2}}{2}\right\} \int_{-l_{1}}^{l_{2}} d\tau \int_{-\alpha_{0}}^{\alpha_{0}} d\alpha_{1} \int_{-\alpha_{0}}^{\alpha_{0}} d\alpha_{2}$$

$$\times \delta(k_{2x} - k_{1}^{0}\alpha_{1} - k_{1}^{0}\alpha_{2}) \exp\left\{\frac{il_{2}(k_{2}e^{\beta} + k_{1}^{0}\alpha_{1} + k_{1}^{0}\alpha_{2})^{2}}{2k_{2}e^{e}} + \frac{ik_{1}^{0}\tau}{4}[(\alpha_{1} - \alpha_{2})^{2} - 4\beta(\alpha_{1} + \alpha_{2})]\right\}.$$
(23)

It follows directly from (23) that $A_2(k_{2X}, l_2)$ can assume values differing noticeably from zero only if the following conditions are simultaneously satisfied:

$$k_{2x} = k_1^{0} \alpha_1 + k_1^{0} \alpha_2, \qquad (24)$$

$$(\alpha_1 - \alpha_2)^2 = 4\beta(\alpha_1 + \alpha_2). \qquad (25)$$

For fixed k_{2x} , relations (24) and (25) impose conditions on the fundamental-radiation wave-vector projections effectively participating in the generation of the harmonic plane wave, whose wavevector projection on the x axis is equal to k_{2x} .

Here (24) is the ratio of the projections of the wave vectors on the x axis, and (25) (with (17) taken into account) is the same for the z axis. In place of (24) and (25) we can write the vector relation

$$\mathbf{k}_{1}^{(1,0)} + \mathbf{k}_{1}^{(2,0)} = \mathbf{k}_{2}^{e},$$
 (26)

which can be called the synchronism condition. Condition (26) is satisfied only for one-dimensional interaction $(\mathbf{k}_1^{(1,0)} \| \mathbf{k}_1^{(2,0)})$ if $k_{2\mathbf{X}} = 0$, if $k_{2\mathbf{X}} \neq 0$ and $(\mathbf{k}_1^{(1,0)} \neq \mathbf{k}_1^{(2,0)})$ for two-dimensional interaction (see Fig. 1).

From (24) and (25) we can determine the angles α_1 and α_2 at which the fundamental waves satisfying the condition (26) propagate:

$$\alpha_{1} = k_{2x} / k_{2}^{e} \pm \sqrt{2\beta k_{2x} / k_{2}^{e}}, \quad \alpha_{2} = k_{2x} / k_{2}^{e} \mp \sqrt{2\beta k_{2x} / k_{2}^{e}}.$$
(27a)

It follows from (27a) that real values of $\alpha_{1,2}$ correspond only to positive k_{2X} . The latter denotes that effective generation of the second harmonic in the focused beam occurs only on one side of the one-dimensional synchronism direction. Therefore the form of the angular spectrum of the second harmonic differs essentially from the angular spectrum of the fundamental radiation; the symmetrical spectrum A_1 (see (9)) gives way to an asymmetrical spectrum A_2 . The limiting value of k_{2X} for which (26) can still be satisfied in a beam with divergence α_0 is determined by the relation

$$k_{2x}^{(\lim)}/k_{2^e} = \alpha_0 + \beta - \sqrt{\beta(2\alpha_0 + \beta)}.$$
 (27b)

We now turn to a detailed study of a spatial structure and of the power of the second harmonic. Integrating (23) we obtain

$$A_{2}(k_{2x}, l_{2}) = \frac{-i2 \sqrt{2\pi} \gamma P_{1}}{c b a_{0} k_{1^{0}}} \exp\left(\frac{ik_{2x}^{2} l_{2}}{2k_{2}^{e}} + i\beta k_{2x} l_{2}\right)$$

$$\times \left\{ \int_{0}^{(k_{1}^{0} l_{1})^{1/2}} [C(\eta_{1}) - iS(\eta_{1})] \exp\left(\frac{i\beta k_{2x} \eta^{2}}{k_{1^{0}}}\right) d\eta + \int_{0}^{(k_{1}^{0} l_{2})^{1/2}} [C(\eta_{1}) + iS(\eta_{1})] \exp\left(\frac{-i\beta k_{2x} \eta^{2}}{k_{1^{0}}}\right) d\eta \right\}; \quad (28a)$$

$$A_{2}(k_{2x}, l_{2}) = 0 \text{ for } |k_{2x}| / k_{2}^{e} > a_{0}. \quad (28b)$$

Here $C(\eta_1)$ and $S(\eta_1)$ are the Fresnel integrals, and

$$\eta_1 = \langle (\alpha_0 - |k_{2x}| / k_2^e) \eta.$$

In order to illustrate the main features of the second-harmonic generation process in a focused beam, we shall consider separately two limiting cases: focusing of the beam with the aid of lenses with long focus ($z_f \gg L$) and short focus ($z_f \ll L$).

5. CHARACTERISTICS OF SECOND HARMONICS EXCITED BY A LONG-FOCUS CYLINDRICAL LENS

The condition $z_{\rm f}\gg L$ is equivalent (see (11)) to the relation

$$\alpha_0 \gamma \overline{k_1^0 L} \ll 1. \tag{29}$$

Using the expansion of the Fresnel integrals for small values of the argument

$$C(\eta_{1}) = \sqrt{\frac{2}{\pi}} \left(\eta_{1} - \frac{\eta_{1}^{5}}{5 \cdot 2!} + \dots \right),$$

$$S(\eta_{1}) = \sqrt{\frac{2}{\pi}} \left(\eta_{1}^{3} - \frac{\eta_{1}^{7}}{7 \cdot 3!} + \dots \right),$$
(30)

we can calculate approximately the integral (28)

$$A_{2}(k_{2x}, l_{2}) = \frac{2\gamma P_{1}}{c b \alpha_{0} \beta k_{2x}} \left(\alpha_{0} - \frac{|k_{2x}|}{k_{2}^{e}} \right) (1 - \exp\{i\beta k_{2x}L\}).(31)$$

Using (31), we can calculate the amplitude of the harmonic at the point of observation $A_2(x, z)$ (see formula (21)) and the total power of the second harmonic at the output of the nonlinear crystal

$$P_{2} = \frac{cb}{4\pi} \cdot 2\pi \int_{-\infty}^{\infty} A_{2}(k_{2x}, l_{2}) A^{*}(k_{2x}, l_{2}) dk_{2x}.$$
(32)

From (31) we easily see that the expressions for $A_2(x, z)$ and P_2 are determined essentially by the relation between the wave divergence α_0 and the anisotropy angle β .

When $\alpha \ge \beta$, and consequently (see (29)) when $2\beta\alpha_0 k_1^0 L \ll 1$, we have

$$P_{2}' = 8\gamma^{2} P_{1}^{2} \alpha_{0} k_{1}^{0} L^{2} / 3cb, \qquad (33)$$

and the distribution of $A_2(x, z)$ remains homogene-

ous in x. When $\beta \gg \alpha_0$, such that $2\beta \alpha_0 k_1^0 L \gg 1$, we have

$$P_{2}'' = 4\pi\gamma^{2}P_{1}^{2}L / cb\beta.$$
 (34)

The quantity $A_2(x, z)$ can be calculated in this case by the stationary-phase method (the observation is usually carried out at distances sufficiently far from the crystal $z - l_2 \gg z_f$). Depending on the observation angle $\theta = (x - \beta L/2)/z$ we obtain for $A_2(x, z)$

$$|A_2(x, z)|^2 = \frac{16\pi\gamma^2 P_1^2 \sin^2\beta k_0 \theta L}{c^2 b^2 k_0 \beta^2 \theta^2 z}.$$
 (35)

Formulas (33) and (34) show that $z_f \gg L$ when the dominant role in the second-harmonic generation process is assumed by one-dimensional interactions. Indeed, formula (33) has the same structure as the formula for the power of the harmonic generated by a plane wave propagating in the onedimensional synchronism direction $P_2^{(pl)}$ = $4\pi\gamma^2 P_1^2 L^2/c\sigma$, where σ is the area of the fundamental-radiation beam.

Formulas (34) and (35) are similar to the formulas derived in ^[2,3] for a weakly diverging beam, in which the generation of the second harmonic is produced by one-dimensional interactions along each of the rays.¹⁾ In analogy with [2, 3] we can introduce here, too, the concept of the coherentinteraction length $L_c = 2\pi/\beta \alpha_0 k_1^0$. Then the condition for the applicability of (33) is $L/L_c \ll 1$, and deviations from the conditions of one-dimensional synchronism over the cross section of the focus are insignificant, while for (34) and (35) we have $L/L_c >> 1$ (deviations from the conditions of onedimensional synchronism over the section of the focus are appreciable). With the aid of (33) and (34) we can determine the energy gain resulting from focusing the main radiation, by comparing P'_2 and P''_2 with the powers of the harmonics generated over the same length by an unfocused beam of area $\sigma = b^2$. The quantity P₂' should obviously be compared with $P_2^{(pl)}$, and P_2'' with the power of the second harmonic generated by a beam having a divergence α_0 and an area b². We have

$$\eta' = \frac{P_2'}{P_2^{(p1)}} = \frac{2}{3} \frac{b}{x_f}, \quad \eta'' = P_2 \left[\frac{4\pi \gamma^2 P_1^2}{cb^2} LL_c \right]^{-1} = \frac{b}{x_f}.$$
 (36)

Thus, the gain due to the focusing is determined when $z_f \gg L$ only by the increase in the field intensity of the focus; calculations of the characteristics of the second harmonics can be carried out in this case with the aid of the formulas derived in the geometric-optics approximation by substituting in them bx_f in lieu of b².²⁾

6. EXCITATION OF SECOND HARMONICS WHEN FOCUSING A BEAM WITH A SHORT FOCUS LENS IN THE MIDDLE OF THE CRYSTAL

When a beam is focused by a short-focus lens, the length of the crystal exceeds the size of the focal spot, so that the following relation is satisfied:

$$\alpha_0 \sqrt[7]{k_1^0 L} \gg 1. \tag{37}$$

Without loss of generality, to simplify the final expressions, we can confine ourselves to an examination of the focusing of a beam in the center of the crystal, that is, we can put $l_1 = l_2$. Then inequality (37) will be satisfied for either l_1 or l_2 , from which it follows that the focal spot is sufficiently far from both the front and the rear boundaries of the crystal. When condition (37) is satisfied we can obtain for the integral (28) an asymptotic equation that makes it possible to express $A_2(k_{2x}, l_2)$ in the following manner:

$$A_{2}(k_{2x}, l_{2}) = \frac{-i2\gamma P_{1} \gamma 2\pi k_{1} \overline{l_{2}}}{c b a_{0} k_{1}^{0}} \exp\left\{\frac{ik_{2x} l_{2}}{2k_{2}^{c}} + i\beta k_{2x} l_{2}\right\} \\ \times \left[\Phi\left(\sqrt{\beta k_{2x} l_{2}}\right) + O\left(\frac{1}{\sqrt{k_{1} \overline{l_{2}}} (a_{0} - |k_{2x}|/k_{2}^{c})^{2}}\right)\right], \quad (38)$$

where the amplitude distribution function is

$$\Phi(\sqrt{\beta k_{2x}l_2}) = \frac{C(\sqrt{\beta |k_{2x}|l_2}) \pm S(\sqrt{\beta |k_{2x}|l_2})}{\sqrt{2\beta |k_{2x}|l_2/\pi}}$$
$$-\begin{cases} \left[\frac{2}{\pi}\beta |k_{2x}|l_2\right]^{-1/2} & \text{for } \frac{k_{2x}}{k_2^e} > \alpha_0 + \beta - \sqrt{\beta(2\alpha_0 + \beta)} \\ 0 & \text{for } \frac{k_{2x}}{k_2^e} < \alpha_0 + \beta - \sqrt{\beta(2\alpha_0 + \beta)} \end{cases}$$
(39)

In formula (39) the plus sign should be taken for positive k_{2x} and the minus sign for negative k_{2x} . The last term of (38) indicates the order of magnitude of the quantities discarded when evaluating the integral.

Using (38) and (39) we can calculate the inten-

¹⁾We note that by using (28) we can also consider, in a manner more rigorous than used $in^{[2,3]}$, the generation of harmonics in a weakly diverging beam exciting a nonlinear crystal situated far from the phase center of the wave. We can obtain the formulas derived $in^{[2]}$ by assuming that the signs of l_1 and l_2 are the same and that $l_2 = L + l_1$ (L $\ll l_1$), and by integrating (28).

²⁾For a diverging beam with divergence other than a_0 and equal to a_1 , we get $\eta \, " \approx a_1 b/a_0 x_f$ and at $a_1/a_0 \ll 1$ the gain due to focusing can decrease noticeably: $\eta \, " = 1$ when $a_1 = \lambda/b$, i.e., in the case of diffractive divergence.



sity of the second harmonic at the point of observation and the total power of the second harmonic. Since condition (37) is satisfied we can use the method of stationary phase to calculate the integral (21) and then the quantity $|A_2(x, z)|^2$ takes the form

$$|A_{2}(x,z)|^{2} = \frac{8\pi^{2}\gamma^{2}P_{1}^{2}l_{2}k_{0}}{c^{2}b^{2}\alpha_{0}^{2}zk_{1}^{0}} |\Phi(\overline{\gamma^{2}\beta l_{2}k_{0}\theta})|^{2}, \qquad (40)$$

where $\theta = (x - \beta l_2)/z$ (if k_1^0 and k_0 differ greatly, then z must be replaced by $z_1 = z - l_2(k_1^0 - k_0)/k_1^0$). Thus, the character of the spatial structure of the second-harmonic radiation is determined by the form of the function Φ . Figure 3 (curve 1) shows a plot of $|\Phi|^2$ as a function of the reduced value of the angle

$$\tilde{\theta} = 2\beta k_0 \theta L / \pi.$$

The same figure shows for comparison a plot of the spatial structure of the second harmonic, generated by a diverging beam in which only onedimensional interactions occur³⁾

$$|A_2(x,z)|^2 \sim (\beta k_0 \theta)^{-2} \sin^2 \beta k_0 \theta L$$

(curve 2); the central maximum of curve 2, which contains more than 90% of the second-harmonic power, spans an angle $\tilde{\theta}_0 = 4$.

Comparison of curves 1 and 2 shows that their forms differ most strongly in the region of positive k_{2X} , obviously because of the contribution of the two-dimensional interactions when a shortfocus lens is used (see formula (27a)). On the other hand, in the region of negative k_{2X} the generation of the harmonic is due to one-dimensional FIG. 3. Spatial structure of radiation of second harmonic (curve 1) excited at the output of a nonlinear crystal by a short-focus lens. Ordinates – square of the modulus of the function Φ , proportional to the intensity of the second harmonic; abscissa – normalized value of the observation angle $\tilde{\theta} = 2\beta k_0 \theta L/\pi$ ($\theta = (x - \beta L/2)z^{-1}$). The figure also shows for comparison the spatial structure of the harmonic generated as a result of one-dimensional interactions in a diverging beam (curve 2), and a plot of θ^{-1} (curve 3).

interactions. The width of the principal maximum of curve 1 is of the order of θ_0 ; with further increase of $\tilde{\theta}$ the intensity of the harmonic decreases approximately like $\tilde{\theta}^{-1}$, and with increasing deviation from $\tilde{\theta} = 0$ the efficiency of the twodimensional interaction decreases. The limiting value θ_{\lim} at which two-dimensional interaction is still observed is determined by formula (39); we note that the value of θ_{\lim} obtained in this manner coincides exactly with the value determined by formula (27b), which has been derived from purely geometric considerations. At small values of the anisotropy angle $\beta \ll \alpha_0$, such that $2\beta \alpha_0 k_1^0 l_2 \ll 1$, the width of the angular spectrum of the second harmonic is determined by the value of the angle α_0 , within the limits of which $\Phi \approx 1$ and consequently

$$|A_{2}(x,z)|^{2} = \begin{cases} \frac{32\pi^{2}\gamma^{2}P_{1}^{2}l_{2}k_{0}}{c^{2}b\alpha_{0}^{2}k_{1}^{0}} & \text{for} & |\theta| < \alpha_{0} \\ 0 & \text{for} & |\theta| > \alpha_{0} \end{cases}; \quad (41)$$

the second-harmonic power is in this case equal to

$$P_2^{\prime\prime\prime} = 16\pi\gamma^2 P_1^2 l_2 / cba_0. \tag{42}$$

It is important to emphasize that in this case the second-harmonic power increases like the first power of the distance l_2 even when $\alpha_0 \gg \beta$, but the reason now is no longer the dispersion of the medium (see (34)), but the decrease in the amplitude of the cylindrical fundamental-radiation wave.

Using (42) and (33) we can compare the powers of the second harmonic at the output of a crystal of fixed length when focusing with a long-focus lens (divergence α_l) and a short-focus lens (α_s)

$$\frac{P_{2^{\prime\prime\prime}}}{P_{2^{\prime}}} = \frac{3z_{\rm f}}{l_2} \frac{\alpha_l}{\alpha_{\rm s}},\tag{43}$$

³)We recall that the harmonic has a similar spatial structure also when a beam is focused by a long-focus cylindrical lens, see formula (35).

in which connection the use of a long-focus lens is preferable.

When $\beta \geq \alpha_0$ the angular aperture of the second harmonic is determined essentially by the width of the maximum of the function Φ , and the total power of the harmonic

$$P_2 = \frac{8\pi^2 \gamma^2 P_1^2}{c b \alpha_0^2 k_0} \left[1 + \frac{1}{4} \ln\left(\frac{\widetilde{\theta}(\lim)}{2}\right) \right]; \qquad (44)$$

the rate of growth of the second harmonic is slowed down further with increasing distance by dispersion, and then becomes logarithmic.⁴⁾

7. CONCLUSION

The results enable us to describe in this manner the most essential features of a harmonic in a focused beam. The qualitative relations that characterize a transition from a short-focus lens to a long-focus lens are in good agreement with data given in ^[3] (see also ^[11]). Focusing such that z_f is close to the length of the nonlinear crystal is apparently optimal from the energy point of view. Although a detailed calculation was made here only for cylindrical waves, the conclusions regarding the relative roles of one-dimensional and two-dimensional interactions is valid also for a spherical wave (in this case the harmonic power gain for $z_f \gg L$ and $\alpha_0 \gg \beta$ in a spherical wave amounts to $\eta = b^2/x_f^2$).

The procedure described makes it possible to analyze also many other nonlinear effects with account of diffraction, such as parametric amplification and induced scattering.

¹ P. D. Maker, R. Terhune, M. Nisenoff, and C. Savage, Phys. Rev. Lett. 8, 21 (1962).

² D. Kleinman, Phys. Rev. **128**, 1761 (1962).
³ S. A. Akhmanov, A. I. Kovrigin, and N. K. Kulakova, JETP **48**, 1545 (1965), Soviet Phys. JETP **21**, 1034 (1965).

⁴S. A. Akhmanov and R. V. Khokhlov, Problemy nelineĭnoĭ optiki (Problems of Nonlinear Optics), AN SSSR, 1964.

⁵G. D. Malyuzhinets, UFN **69**, 321 (1959), Soviet Phys. Uspekhi **2**, 749 (1960).

⁶ G. D. Malyuzhinets and L. A. Vainshtein,

Radiotekhnika i élektronika **6**, 1247 and 1489 (1961). ⁷ V. M. Faĭn and Ya. I. Khanin, Kvantovaya ra-

diofizika (Quantum Radiophysics), Soviet Radio Press, 1965.

⁸ A. P. Sukhorukov and R. V. Khokhlov, Vestnik, Moscow State University (in press).

⁹ A. Sommerfeld, Optics (Russ. Transl.), IIL, 1953.

¹⁰ M. Born and E. Wolf, Principles of Optics, London, 1959.

¹¹ R. Miller and A. Savage, Phys. Rev. **128**, 2175 (1962).

Translated by J. G. Adashko 63

⁴)It is interesting that a similar growth rate is possessed also by a spherically diverging wave when $a_0 >> \beta$.