INFLUENCE OF THE FINITE APERTURE OF A LIGHT BEAM ON NONLINEAR EFFECTS IN AN ANISOTROPIC MEDIUM

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The spatial structure of the second harmonic radiation generated by a light beam of finite aperture in an anisotropic medium is investigated. Special attention is devoted to nonlinear effects near the so-called synchronism direction. A method is shown for determining the coherent interaction length in second harmonic generation by a diverging beam of the fundamental radiation. Some features of the nonlinear interaction of waves in focused beams are noted. Effects connected with changes of the crystal anisotropy parameter are investigated.

1. INTRODUCTION. MODELS OF THE FUNDAMENTAL BEAM

THE results of the first experiments in nonlinear optics, performed in 1961 and 1962,[1,2] were generally interpreted using the theory of interactions between unmodulated plane waves that was developed by several authors.[3-5] Although this theory satisfactorily describes the general features of the observed phenomena, a number of effects do not fit into the proposed scheme. It has been shown, particularly,[6-8] that even a relatively small degree of nonmonochromaticity of the fundamental radiation can produce appreciable effects in experiments on the generation of optical harmonics, of sum and difference frequencies, etc. The same can be said regarding the influence of slight divergence of a fundamental beam. We shall now discuss this effect in some detail.

Second harmonic generation (SHG) in a nonlinearly polarized medium when the efficiency of energy transfer from a given plane fundamental beam to the second harmonic is only a few percent can be described by[4,7]

$$p_2 = \beta p_1^2 \Delta^{-2} \sin^2 (\Delta l / 2). \tag{1}$$

Here p_1 and p_2 are the intensities of the fundamental beam and the second harmonic, β is a coefficient determined by the nonlinear properties of the medium, l is the length of the nonlinear medium, and Δ is a quantity characterizing the dephasing of the interacting waves:

$$\Delta = k_2 - 2k_1 = (2\pi / \lambda_2)(n_2 - n_1). \tag{2}$$

Here λ_2 is the wavelength of the second harmonic; n_1 and n_2 are the refractive indices for the fundamental and the second harmonic.

Utilizing (1) and (2), we introduce the concept of a length of coherent interaction between plane fundamental and second harmonic waves:

$$l_{k0} = \lambda_1 / 4(n_2 - n_1) \tag{3}$$

(here λ_1 is the fundamental wavelength). The quantity l_{k_0} is directly related to energy; for $l < l_{k_0}$ the second harmonic intensity increases monotonically with distance, while for $l > l_{k_0}$ it oscillates. In a given fundamental field the second harmonic intensity generated by a plane wave does not exceed

$$p_{2\,max} = \beta p_1^2 l_{k0}^2. \tag{4}$$

A plane wave is, of course, an extremely crude model. Light beams utilized in nonlinear optical experiments have a finite aperture; therefore care must be exercised in applying concepts developed for plane waves to the interpretation of the experimental results. The character of nonlinear interactions in a finite-aperture beam depends on the beam structure and the dispersive properties of the nonlinear medium. The two most important features are: 1) Nonlinear interactions of intersecting rays can occur in a finite-aperture beam, and 2) the dispersive properties of a nonlinear medium may not be identical for different rays of a finite-aperture beam, particularly when nonlinear effects in anisotropic media are investigated.

It has been shown by both theoretical calculations and experiments,[1,2,7,9] that the foregoing factors are practically unimportant when nonlinear effects are observed in unfocused laser beams, but that the phase velocities of the interacting waves do not coincide for a single ray. This is the situation in experiments on the generation of optical harmonics by an unfocused fundamental beam in optically isotropic media and in anisotropic media exhibiting relatively small birefringence. Here even for a beam of finite divergence the variation of Δ over a cross section can be neglected and the theory developed for plane waves can be used, particularly the concept of a coherence length represented by (3). The second harmonic intensity can be calculated by multiplying (1) by the effective beam area. The correctness of this procedure when Δ is practically identical and nonvanishing for all rays of the fundamental beam [$\Delta(\theta,\varphi)/\Delta(0,0) \approx 1$ for the entire beam; here $\Delta(0,0)$ is the value of Δ on the beam axis, while θ and φ are the polar angles] was confirmed experimentally in [2].

The situation is different when the so-called synchronism direction exists along one of the rays with $\Delta(\theta,\varphi) = 0$; we shall hereafter consider the most interesting case of a negative uniaxial crystal (Fig. 1). Here even small divergence of the fundamental radiation can strongly affect SHG. To show this, we consider the generation of the harmonic in a fundamental beam emerging from a spherical cavity resonator,[1)] and possessing the angular divergence $2\alpha_1$. There are two important factors for this model of the fundamental beam in connection with SHG. First, unlike the plane wave case, the given field of the fundamental decreases with distance; secondly, Δ is not identical for different rays of the beam [see Eq. (2)]. However, if the phase center of the fundamental wave is located at a sufficient distance from a nonlinear medium and the length of the nonlinear medium is relatively small ($l/R_0 \ll 1$, where l is the length of the nonlinear crystal and R_0 is the distance from the phase center), in the approximation of geometric optics the considered fundamental beam can be represented as a bundle of rays with harmonic generation along each ray described by Eqs. (1)—(3).[2)] Since the refractive index surfaces are symmetric about the optic axis, Δ depends only on θ. Near the synchronism direction we have

$$\Delta = K\delta\theta \tag{5}$$

where $\delta\theta$ is the deviation from the synchronism angle θ_S and K is a constant determined by the dispersive properties of the crystal.

The second harmonic intensity distribution will, consequently, assume the form of bands defined by (1) and (5) with > 90% of the total radiation included in the angle $2\alpha_2$ [the central maximum of the function $\Delta^{-2}\sin^2(\Delta l/2)$] where

$$\alpha_2 = 2\pi / lK. \tag{6}$$

The dependence of second harmonic intensity on φ is contained in the dependences of the coefficient β ($\beta \sim \sin 2\varphi$)[2,7] and $p_1(\varphi)$. The angle $2\delta\varphi$ containing most of the harmonic intensity for a slightly diverging fundamental beam can be assumed to equal $2\alpha_1$.

Assuming that the entire second harmonic intensity is contained in the central maximum of cross section σ_2, we have

$$P_2 = \sigma_2 p_2 = n_2 \frac{c}{8\pi}\sigma_2 E_2^2 = n_2 \frac{c}{8\pi}\sigma_2 \beta E_1^4 l^2$$
$$= \left(\frac{c}{8\pi}\right)^{-1} \frac{n_2}{n_1}\frac{P_1^2}{\sigma_1}\frac{\sigma_2}{\sigma_1} l^2. \tag{7}$$

Here E_1 and E_2 are the field strengths, P_1 and P_2 are the total fundamental and harmonic intensities, σ_1 is the cross section of the fundamental beam, and c is the velocity of light. Obviously,

$$\sigma_2 / \sigma_1 = \alpha_2 / \alpha_1 = 2\pi / lK\alpha_1,$$

and, consequently, for $\sigma_2 \geq \sigma_1$ the second harmonic intensity

$$P_2 = \left(\frac{c}{8\pi}\right)^{-1} \frac{n_2}{n_1}\frac{P_1^2}{\sigma_1} l^2$$

increases as the square of distance, while for $\sigma_2 < \sigma_1$ we have

$$P_2 = \left(\frac{c}{8\pi}\right)^{-1} \frac{n_2}{n_1}\frac{P_1^2}{\sigma_1}\frac{2\pi}{K\alpha_1} l$$

with a linear dependence on distance.

The length $l = l_{K\alpha}$, for which $\sigma_2 = \sigma_1$, can be called the coherent interaction length for a diverging beam:[3)]

$$l_{k\alpha} = 2\pi / K\alpha_1. \tag{8}$$

We note that for the given model the maximum

[1)]This is supported by recently published experimental investigations[10,11] of the spatial structure of radiation from solid state lasers.

[2)]We note that in this case, when variations of the fundamental field strength cannot be neglected, harmonic generation in a spherical wave must be considered. The corresponding calculation shows that here, unlike (1), the intensity increases logarithmically with distance for small l. We intend to make a separate, more detailed, examination of this question.

[3)]This definition of the coherence length is convenient for analyzing experimental data. Also, $l_{K\alpha}$ agrees in order of magnitude with the quantity introduced by Kleinman,[9] who averaged (1).

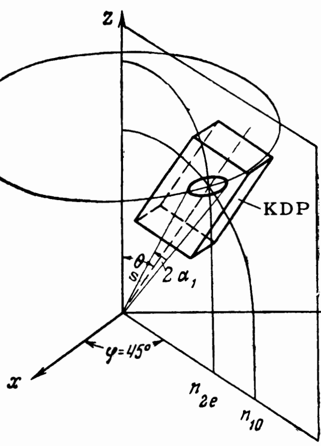

FIG. 1. Intersection of surfaces of the refractive indices n_1 and n_2 in a negative uniaxial crystal by the zx′ plane. $\theta = \theta_S$ corresponds to the direction in which the fundamental ordinary wave and second harmonic extraordinary wave have equal phase velocities.

efficiency for frequency doubling is, obviously,

$$\eta = p_1\sigma_2 / p_1\sigma_1 = \alpha_2 / \alpha_1 = l_{k\alpha} / L_2, \quad (9)$$

where $L_2 = (k_1 \chi E_1)^{-1}$ is the length in which the second harmonic intensity attains equality with the fundamental intensity.[3,4,7] Here χ is the corresponding element of the nonlinear polarization tensor. It follows specifically from the last equation that at high efficiencies we have $\eta \sim P_1^{1/2}$ for a diverging beam (for $l \ll l_{k\alpha}$ and $P_2 \ll P_1$ we have $\eta \sim P_1$).

We emphasize again that Eqs. (5)—(9) describe one-dimensional interactions in a paraxial beam of finite aperture. If interactions of intersecting beams are also possible, the spatial structure of the second harmonic will be changed, as a general rule. The SHG in any direction (θ, φ) is now associated not only with the combining of fundamental photons moving in the (θ, φ) direction, but also with the combining of photons moving at an angle $(\theta + x, \varphi + y)$. If the fundamental radiation excites extraordinary waves in a crystal [by means of the interaction $\gamma_0(\omega) + \gamma_0(\omega) \rightarrow \gamma_e(2\omega)$; see Fig. 1], the directions of the mixed waves are symmetric about the given direction (θ, φ), and in the given field approximation the second harmonic intensity is

$$p_2(\theta, \varphi) = \int \beta p_1^2(x, y) \frac{\sin^2[\Delta(x, y, \theta, \varphi) l/2]}{\Delta^2(x, y)} dx\, dy, \quad (10)$$

where x and y are the deviations of the mixed fundamental ray directions from the given direction, and

$$\Delta(x, y, \theta, \varphi) = k_2(\theta, \varphi) - 2k_1\Phi(x, y).$$

In a uniaxial crystal the angular structure of the second harmonic is practically independent of the angle φ; we can therefore confine ourselves to an analysis of the interaction in the plane of the optic axis. In this case $\Delta(\theta, x) = k_2(\theta) - 2k_1 \cos x$, thus showing that the vanishing of $\Delta(\theta, x)$ and, therefore, the occurrence of combining interactions are possible in the region $\theta > \theta_S$, where $k_2 < 2k_1$. For small angles the following expression is valid for $\Delta(\theta, x)$:

$$\Delta(\theta, x) = K\delta\theta + 2k_1x^2. \quad (11)$$

The spatial structure changes considerably by comparison with the foregoing case of one-dimensional interactions when $\Delta(\theta, x) = 0$ for directions along which $\sin(K\delta\theta l/2) = 0$. The minimum beam divergence for which this condition is fulfilled will be, in accordance with (6) and (11),

$$\alpha_{min} = (\lambda_1 / n_1 l)^{1/2} \quad (12)$$

(which for a potassium dihydrogen phosphate (KDP) crystal 2 cm long gives $\alpha_{min} = 5 \times 10^{-3}$ rad).

We present here the results obtained in an experimental investigation of the way in which the aperture of a fundamental beam affects SHG along the synchronism direction. The spatial structure of the harmonic is obviously most sensitive to aperture effects. A careful analysis of this structure yields information regarding the structure of the fundamental beam, the relative part played by the interaction of intersecting rays etc. The experiments were performed with fundamental beams of different configurations (focused and unfocused) and with crystals of different lengths.

2. EXPERIMENT

Figure 2 represents the experimental arrangement designed for investigating the spatial structure of the second harmonic. A laser beam formed with external mirrors impinges on a KDP crystal cut in the form of a prism and fastened to a special rotating device enabling orientation of the crystal

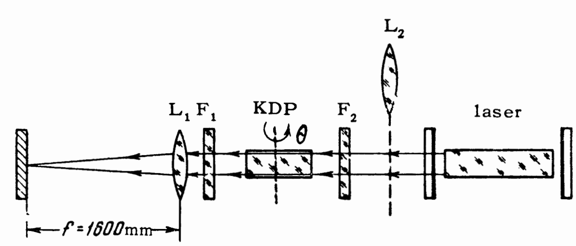

FIG. 2. Block diagram of experimental setup designed to investigate the spatial structure of the second harmonic.

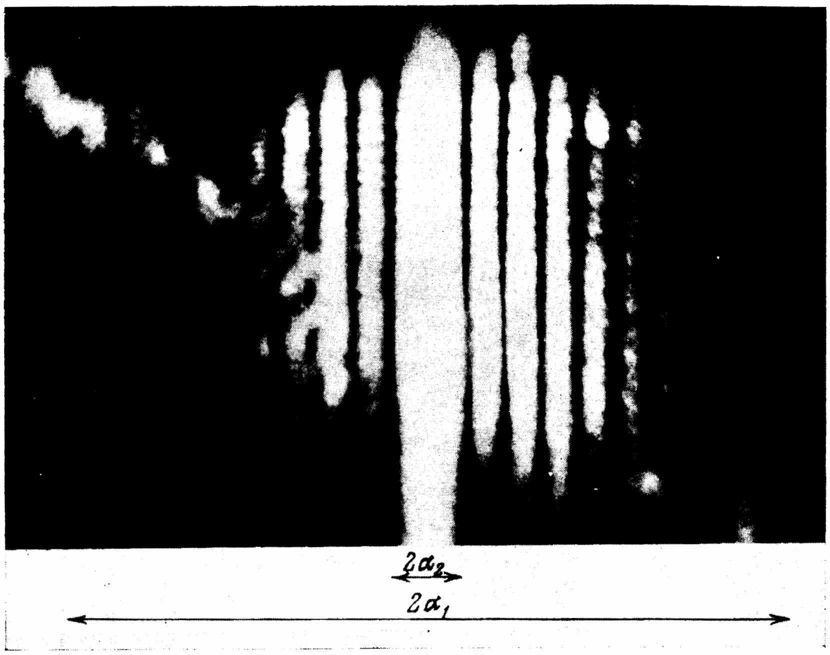

FIG. 3. Photograph of cross section of second harmonic ray excited in a KDP crystal 2 cm long. The crystal was oriented to have the fundamental beam direction along the synchronism direction. The corresponding angular scale is shown in Fig. 4.

to within 20″. The second harmonic traverses the filter F_1 which absorbs the fundamental frequency, and passes through the objective lens L_1 of focal length f = 1600 mm. The long-focus objective resolves the angular structure of the second harmonic to within 1″. The radiation is photographed on a plate located in the focal plane (at f) of the objective.

Figure 3 represents a typical spatial structure of a second harmonic ray generated by an unfocused ruby laser beam in a KDP crystal of length $l = 2$ cm. The divergence of the fundamental beam was 20′. Figure 4 represents the photometric analysis of Fig. 3. The experiment shows that the number of dark and bright bands in the cross section of a harmonic ray depends essentially on the divergence of the fundamental beam. When this divergence was reduced to 1′—2′ only one band, instead of 10—12 bands (as in Fig. 3), was observed when a KDP crystal 2 cm long was used.

In order to prove that the minima and maxima of second harmonic intensity are associated with the dispersive properties of the crystal and are not induced by any diffractive effects, we varied the dispersive properties artificially by means of an external static electric field. This external field was applied to the lateral faces of the crystal, which was 1.6 cm thick. Figure 5 shows that the spatial structure of the second harmonic changes when the static field is applied. The photographs also show that effective modulation of the harmonic results from variation of the crystal anisotropy parameters.

A final experimental run was performed using focused fundamental beams and different lenses with f varying from 50 to 2 cm. The laser crystal diameter was 1 cm. With each lens the spatial structure of the harmonic was registered for different positions of the focus with respect to the KDP crystal; the focus was located either within, behind, or on the surface of the crystal, or between the crystal and the laser. In those instances where the focus was outside the crystal the spatial structure of the harmonic was very similar to that shown in Fig. 3; the angular separation of the alternating bright and dark bands was the same as for an unfocused beam.

Considerable differences resulted when the fundamental beam was focused inside or on the surface of the crystal. Unlike the case of an unfocused beam, the spatial structure of the harmonic now becomes asymmetric with respect to the plane of θ_S.

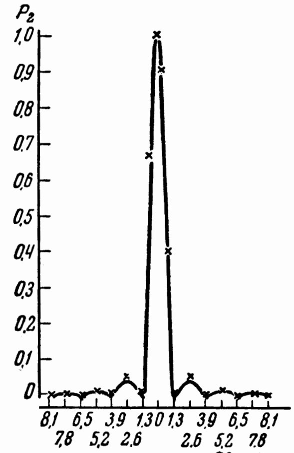

FIG. 4. Distribution of second harmonic intensity in the ray cross section of Fig. 3. The curve was plotted from Eq. (1).

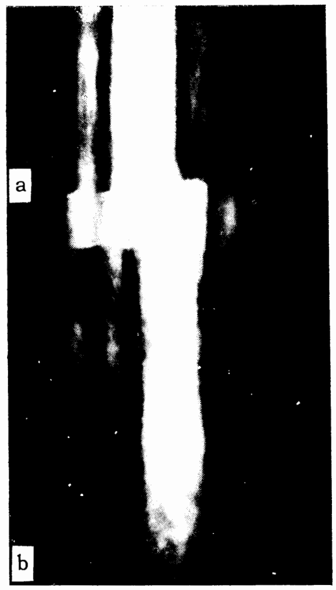

FIG. 5. Photograph of cross section of second harmonic ray when an electric field is applied to the crystal. $a - V = 0$ kV, $b - V = +8$ kV

For $\theta < \theta_S$ a relatively clear pattern of bands remains, and their angular divergence is close to that obtained with an unfocused beam. For $\theta > \theta_S$, in addition to the band pattern there appears a continuous background corresponding to the interaction of intersecting beams and increasing in strength as f is reduced. Figure 6, a typical photograph of the foregoing, was obtained with an f = 4 cm lens; in this case the focus was inside a KDP crystal 3 cm long.

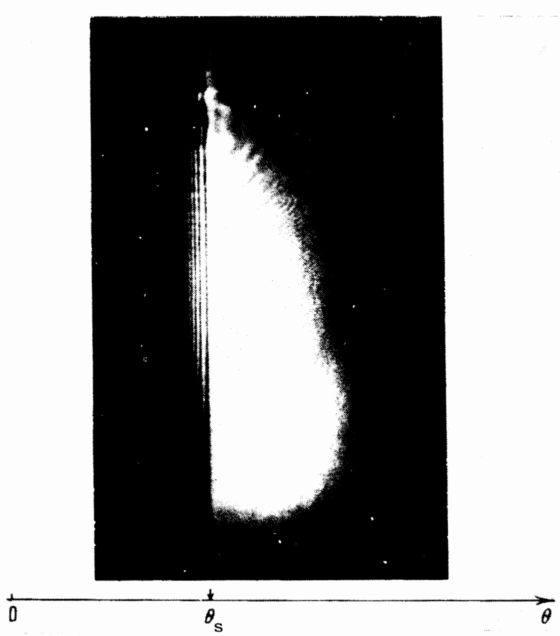

FIG. 6. Photograph of cross section of second harmonic ray excited in a KDP crystal 3 cm long, excited by a focused fundamental bean (f = 4 cm). The focus was located inside the crystal. The angles between the bands for $\theta < \theta_S$ are close to the angles calculated from Eq. (1).

3. DISCUSSION

The angular structure of a second harmonic ray (Fig. 3) shows that an unfocused laser beam is very similar to a paraxial beam emitted by a point source. Indeed, the clear sequence of bright and dark bands observed experimentally and their satisfactory description by Eq. (1) (Fig. 4) indicates that there is no appreciable contribution from intersecting rays in SHG. The experimental locations of the dark and bright bands and those calculated from (1) and (5) agree very accurately [for the interaction $\gamma_0(\omega) + \gamma_0(\omega) \rightarrow \gamma_e(2\omega)$ in a KDP crystal with $\lambda_1 = 6943$ Å and $K = 0.84 \times 10^4$ cm^{-1}].

We now present the angles calculated from (1) and (5), and those observed experimentally, of the bands in the cross section of a second harmonic ray excited by a ruby laser ray in a KDP crystal 2 cm long; N is the number of the band:

N	1	2	3	4	5
Theory	1.33′	2.66′	4.00′	5.33′	6.65′
Experiment	1.39′	2.73′	4.13′	5.53′	6.87′

A direct calculation of the number m of bright bands that can be fitted in the aperture of the fundamental beam enables us to determine the coherence length $l_{k\alpha}$ from the crystal length. The locations of the dark bands are given by

$$K\delta\theta_n l / 2 = N\pi \tag{13}$$

and therefore $l_{k\alpha} = 2l/(m+1)$. It is thus indicated that the length of the KDP crystal for the experiment represented by Fig. 3 is approximately 10 times $l_{k\alpha}$. In the ruby laser experiments $l_{k\alpha}$ varied from 1 to 3 mm [for the interaction $\gamma_0(\omega) + \gamma_0(\omega) \rightarrow \gamma_e(2\omega)$].[4)]

The results obtained when an external static field was applied are in good agreement with the theory. We calculated the band shift resulting from the changes of refractive indices that were induced by the field. A KDP crystal was cut along the direction in which maximum SHG occurs, so

[4)]It must be remembered that for a given aperture α, the value of $l_{k\alpha}$ is not identical in different nonlinear interactions. In addition to the interaction $\gamma_0(\omega) + \gamma_0(\omega) \rightarrow \gamma_e(2\omega)$, in some nonlinear crystals the combining interaction $\gamma_0(\omega) + \gamma_e(\omega) \rightarrow \gamma_e(2\omega)$ is also possible.)[12] For the visible spectrum of some crystals K pertaining to the latter reaction is approximately one-third of its magnitude for $\gamma_0(\omega)+\gamma_0(\omega) \rightarrow \gamma_e(2\omega)$.

that the fundamental and second harmonic rays were propagated in a plane forming an angle of 45° with the crystallographic axes. (Here $\beta \sim \sin 2\varphi$ becomes maximal; see Fig. 1.) For this crystal orientation the variations (Δn_{1o} and Δn_{2e}) of the refractive index n_{1o} for the ordinary wave of frequency ω_1 and of n_{2e} for the extraordinary wave at 2ω are

$$\Delta n_{1o} = -{}^1/_2 n_{1o}^3 r_{63} E_z,$$

$$\Delta n_{2e} = {}^1/_2 (n_{2o}^{-2} \cos^2\theta + n_{2e}^{-2} \sin^2\theta)^{-3/2} r_{63} E_z \cos^2\theta, \quad (14)$$

where E_z is the electric field along the z axis, and θ is the angle between the ray and the z axis.

The experimental conditions permitted application of a 9-kV field to the crystals. We make the following substitutions in (14):

$$E_z = \frac{V}{h} \sin\theta_s, \quad r_{63} = 1.05 \cdot 10^{-9}\,\text{cm/V}, \quad h_{cr} = 1.6\ \text{cm}$$

$\theta_s = 36°$, the synchronism angle for $\lambda_1 = 1.06$, and the corresponding values $n_{1o} = 1.497$, $n_{2o} = 1.512$, and $n_{2e} = 1.47$; we then obtain

$$\Delta n_{1o} = -0.62 \cdot 10^{-5}, \quad \Delta n_{2e} = 0.40 \cdot 10^{-5}.$$

The combined variation of the refractive indices is

$$\Delta n = \Delta n_{2e} - \Delta n_{1o} = 1.09 \cdot 10^{-5}.$$

From (1) a shift amounting to one band corresponds to $\Delta n = \lambda_2 / l$.

Substituting $\lambda_2 = 5300$ Å and $l = 3$ cm, we obtain

$$\Delta n_{\text{I band}} = 1.76 \cdot 10^{-5}.$$

Thus the relative shift of the bands is

$$\Delta n / \Delta n_{\text{I band}} = 0.58.$$

The experimental shift was 0.55 of a band (Fig. 5), which agrees well with the calculation. The slightly lower value can be attributed to imperfect contact between the electrodes and the crystal.[5)]

Some comments are in order concerning the results of experiments with focused beams. To interpret the qualitative picture of the spatial structure in Fig. 6 we must obviously consider the interaction of intersecting rays [see Eq. (10)].

We note that the experimental results obtained with a focused beam depart considerably from Kleinman's theory[9] of SHG in the case of a diffraction-dependent focus. This statement applies particularly to the absence in the region $\theta > \theta_s$ of a distinct band pattern determined by the depth of focus $L_f = 4\lambda_1(2f/d)^2$ utilized in [9]. This last circumstance may result in important corrections to the evaluation of the influence exerted by focusing on nonlinear effects; great care must therefore be exercised in performing calculations based on diffraction theory.

The enhanced harmonic intensity that accompanies focusing is associated with the increase of field strength and with the contribution from the interaction of intersecting beams. An analysis shows that the relative contributions of these two factors depend on the relative lengths of l and $l_{k\alpha}$. The parameters of optimal focusing differ for the regime of the given field and for the regime in which there is an important inverse reaction of the harmonic on the fundamental field.

Our present results have therefore shown that aperture effects very strongly affect SHG close to the synchronism direction. The aperture-dependent coherence length $l_{k\alpha}$ determines to a considerable extent the efficiency of frequency doubling. It must also be emphasized that the quadratic increase of second harmonic intensity with distance when $l \leq l_{k\alpha}$ occurs only when the fundamental line is not broad. In the general case we must take into account not only the variations of refractive indices within the aperture of the fundamental beam, but also their variations within the width of the fundamental line.

In addition to the aperture-dependent coherence length $l_{k\alpha}$ we must introduce the frequency-dependent coherence length $l_{k\omega}$. When a line of finite width is doubled in frequency $l_{k\omega}$ is defined in first approximation by the difference between the group velocities at the frequencies ω and 2ω. It can be shown that

$$l_{k\omega} = \frac{v}{\Delta\omega_1}, \quad \frac{1}{v} = \frac{1}{u_{gr}(\omega)} - \frac{1}{u_{gr}(2\omega)}$$

where u_{gr} is the group velocity at the indicated frequency and $\Delta\omega_1$ is the width of the fundamental line. In our experiments with a ruby laser $l_{k\omega} > l_{k\alpha}$; therefore the aperture effects were dominant. However, for other spectral regions and lasers exhibiting smaller divergence other relations between $l_{k\omega}$ and $l_{k\alpha}$ are possible.

The authors are indebted to V. G. Dmitriev for valuable discussions, and to A. S. Chirkin for a discussion of questions related to frequency doubling in nonmonochromatic waves.

[5)]It should be noted that when an external static field is applied the picture of SHG in a KDP crystal is complicated, as a general rule. In addition to harmonic generation depending on the tensor χ_{ijk} of quadratic nonlinear polarization, we can also find coherent SHG depending on the cubic polarization tensor θ_{ijkl} (if the static field is applied along the optic axis). However, in our experiments this effect was at least three or four orders of magnitude smaller than the SHG associated with the tensor χ.

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