

## THEORY OF SECOND-HARMONIC GENERATION OF LIGHT IN FOCUSED BEAMS

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A theory of second-harmonic generation of light in focused beams is developed by taking vector synchronism into account. The analysis is carried out in the momentum representation with allowance for the possible finite size of the medium taken into account. The two-dimensional problem (cylindrically focused beams) and the three-dimensional problem are considered. The concept of an effective interaction length for vector synchronism in a focused beam is introduced; it can be used for classifying cases of various positions of the focus in the crystal, and various crystal lengths and focusing angles. The efficiency of transformation of light into the harmonic is calculated for the case when the conical lens proposed in<sup>[5]</sup> is employed. The relative efficiencies of various means of second-harmonic production are discussed.

AS is well known, a decisive role in the generation of optical harmonics in crystals (see<sup>[1]</sup>) is played by their dispersion properties: the greatest generation efficiency is attained in the presence of synchronization—equality of the phase velocities of the laser beam and of the harmonic. An intuitive interpretation of this fact is the requirement to satisfy in the generation process not only the energy conservation law  $\omega_2 = \omega_1 + \omega_1'$  but also the momentum conservation law  $\mathbf{k}_2 = \mathbf{k}_1 + \mathbf{k}_1'$ . So far the greatest attention has been paid to the case of the so-called "one-dimensional synchronism," when  $\mathbf{k}_2 \parallel \mathbf{k}_1 \parallel \mathbf{k}_1'$ .

In this paper we consider the more general case of "vector synchronism," when in general  $\mathbf{k}_1$  is not parallel to  $\mathbf{k}_1'$ . The laser beam is assumed to be strictly monochromatic, and the problem is solved in the specified-field approximation. The customarily used qualitative considerations, based on the momentum conservation, can then be extended to obtain quantitative results.

## 1. SOLUTION OF WAVE EQUATION IN THE MOMENTUM (FOURIER) REPRESENTATION

Let us state immediately that the material in this section is not principally new, but we deem it advisable to include it for a better understanding of the main computational part of the paper. The electromagnetic waves  $\mathbf{E}(\mathbf{r}, t)$  are excited by wave sources—the right side of the wave equation—comprising the external polarization  $\mathbf{P}(\mathbf{r}, t)$ . We con-

sider a monochromatic electromagnetic field characterized by a complex vector  $\mathbf{E}(\mathbf{r})$ <sup>1)</sup>.

We go over to the Fourier representation

$$\mathbf{E}(\mathbf{r}) = \int \tilde{\mathbf{E}}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}} d^3\mathbf{k}, \quad (1)$$

and obtain Maxwell's equations in the form

$$(\mathbf{k}^2 \delta_{ij} - k_i k_j - \epsilon_{ij} \omega^2 / c^2) \tilde{E}_j(\mathbf{k}) = 4\pi c^{-2} \omega^2 \tilde{P}_i(\mathbf{k}). \quad (2)$$

One group of problems arises when the field sources are far from the region of interest to us.<sup>2)</sup> In this case we can set the right side equal to zero and replace the effect of real but remote sources by a suitably chosen solution of the free wave equation. This corresponds to specifying the field at the input of the system (say, the field far from the focus at  $r \rightarrow \infty$ ). Then  $\mathbf{P} = 0$  and for free fields the condition of compatibility of the equations in (2) calls for the vanishing of the determinant of their coefficients. This yields the Fresnel equation (see<sup>[2]</sup>, Sec. 77)

$$\det \| k_i k_j - \mathbf{k}^2 \delta_{ij} + \omega^2 \epsilon_{ij} / c^2 \| = 0,$$

which for a specified  $\mathbf{n} = \mathbf{k}/k$  has in general two different solutions and defines the so-called wave vector surface:  $\mathbf{k}^2 = k_1^2(\mathbf{n}), k_2^2(\mathbf{n})$ . To each solution corresponds its own type of polarization of the field

<sup>1)</sup>We have in mind the representation of the real field  $\mathbf{E}(\mathbf{r}, t)$  in the form  $\mathbf{E}(\mathbf{r}, t) = \text{Re} \{ \mathbf{E}(\mathbf{r}) \exp(-i\omega t) \}$ .

<sup>2)</sup>In our case, this will pertain to the field of the laser light.

E:  $\mathbf{e}^1(\mathbf{n})$ ,  $\mathbf{e}^2(\mathbf{n})$ . The general solution of the free wave equation (2) is

$$\tilde{\mathbf{E}}(\mathbf{k}) = \sum_{l=1,2} \mathbf{e}^l(\mathbf{n}) \delta(\mathbf{k}^2 - k_l^2(\mathbf{n})) f_l(\mathbf{n}). \quad (3)$$

This Fourier transform depends essentially only on two independent variables  $\mathbf{n}$ . Such a simplification can be obtained also in the coordinate representation, wherein the function satisfying the monochromatic wave equation can be calculated in the entire volume from its value on a certain surface by using, for example, the Huygens principle. In order to ascertain the input conditions that the solution (3) must satisfy, it is necessary to calculate the asymptotic behavior of the integral (1) as  $r \rightarrow \infty$ . We do this first for the case of an isotropic medium, where  $\epsilon_{ik} = \epsilon \delta_{ik}$ . In this case  $k_l^2 = \epsilon \omega^2 / c^2 = k_0^2$ . Simple calculations (see<sup>[3]</sup> Sec. 124) give for  $r \rightarrow \infty$

$$\mathbf{E}(r\mathbf{n}') \approx \pi i r^{-1} \sum_{l=1,2} \mathbf{e}^l(\mathbf{n}') [f_l(-\mathbf{n}') \exp(-ik_l r) - f_l(\mathbf{n}') \exp(ik_l r)] \quad (3')$$

with the same functions  $f_l(\mathbf{n})$  that are involved in the Fourier transform (3). This establishes the required correspondence.<sup>3)</sup>

In real situations one usually specifies the "incoming" field (i.e., with  $\exp(-ik_0 r)/r$  in one half-space with respect to  $\mathbf{n}'$ ; then the first term in (3) describes the "incoming" waves and the other the waves which have already "passed" beyond  $r \approx 0$ ; the functions  $f_l(\mathbf{n})$  differ from zero here only in one half-space—the one into which the waves are directed. By way of example we note that an ideal (at infinity) spherical wave with an amplitude distribution over the aperture  $E_0(\mathbf{n}) = \pi |f(-\mathbf{n}')| / R$ , converging at the point  $r = 0$ , corresponds to a pure real function  $f(\mathbf{n})$  (accurate to a constant phase).

The second group of problems is characterized by the presence of sources in the region under consideration itself, and at the same time by the absence of waves entering the system<sup>4)</sup>. In this case we have from (2)

$$\tilde{E}_i(\mathbf{k}) = 4\pi\omega^2 c^{-2} \{ \mathbf{k}^2 \delta_{ij} - k_i k_j - (\omega^2 c^{-2} + i\gamma) \epsilon_{ij} \}^{-1} \tilde{P}_j(\mathbf{k}), \quad (4)$$

<sup>3)</sup>Although the correspondence between (3) and (3') as  $r \rightarrow \infty$  is asymptotically correct, the distance  $r$  at which  $\mathbf{E}(r)$  takes the asymptotic value (3') can be larger for strongly non-homocentric beams than that at which  $\mathbf{E}(r)$  is specified experimentally. In this case, to find  $f(\mathbf{n})$  it becomes necessary to extrapolate beforehand the experimentally specified field to larger  $r$ , using the Huygens principle.

<sup>4)</sup>In our case this occurs for the field at the harmonic frequency.

In (4) the reciprocal of the expression in the curly brackets is formally the well-known Cramer's formula for solving systems of linear equations; in these formulas the determinant in the denominator has zeroes precisely at the points corresponding to solutions of the free equation.

The field  $\mathbf{E}(r)$  at  $r \rightarrow \infty$ , corresponding to  $\tilde{\mathbf{E}}(\mathbf{k})$  from (4), can be obtained by calculating the asymptotic value of the integral (1). The latter is determined by the residues at the poles; the infinitesimally small  $\gamma > 0$  leaves only outgoing waves. For an isotropic medium we obtain

$$\mathbf{E}(r\mathbf{n}') \approx 8\pi^2 \omega^2 c^{-2} \sum_l \mathbf{e}^l(\mathbf{n}') (\tilde{\mathbf{P}}(k_l \mathbf{n}') \mathbf{e}^{l*}(\mathbf{n}')) r^{-1} \exp(ik_l r). \quad (4')$$

It is easy to go over from the arbitrary three-dimensional problem to the particular two-dimensional case, assuming that  $\mathbf{E}(r)$  does not contain, for example, a dependence on the coordinate  $z$ . In this case the dependence of all the Fourier transforms on  $k_z$  will have the form  $\delta(k_z)$ . However, it is simplest not to introduce the coordinate  $z$  or the momentum  $k_z$  at all. For the two-dimensional wave equation (Eq. (2) with two-dimensional vector  $\mathbf{k}$ ) we have the following: 1) a solution of the free-wave type (for  $\mathbf{P} = 0$ )

$$\tilde{\mathbf{E}}(\mathbf{k}) = \sum_l \delta(\mathbf{k}^2 - k_l^2) f_l(\varphi) \mathbf{e}^l(\varphi), \quad k_y/k_x = \text{tg } \varphi \quad (5)^*$$

with asymptotic value at  $r \rightarrow \infty$  ( $\tan \varphi' = y/x$ )

$$\mathbf{E}(r, \varphi') \approx (2k_0 r)^{-1/2} e^{-i\pi/4} \sum_l \mathbf{e}^l(\varphi') [f_l(\varphi') \exp(ik_l r) + i f_l(\varphi' + \pi) \exp(-ik_l r)] \quad (5')$$

and 2) solution of the "radiative" type (Eq. (4) with two-dimensional  $\mathbf{k}$ ) with asymptotic value

$$\mathbf{E}(r, \varphi') \approx 2^{1/2} \pi^{3/2} (k_0 r)^{-1/2} 4\pi\omega^2 c^{-2} \sum_l (\tilde{\mathbf{P}}(k_l, \varphi) \mathbf{e}^{l*}) \mathbf{e}^l \times \exp\left(ik_l r + i\frac{\pi}{4}\right). \quad (4'')$$

For the one-dimensional wave equation (Eq. (2) with  $\mathbf{P} = 0$ ) we get

$$\tilde{\mathbf{E}}(k) = \sum_l \delta(k^2 - k_l^2) [f_l^+ \theta(k) + f_l^- \theta(-k)] \mathbf{e}^l, \quad (6)$$

$$\mathbf{E}(x) = \sum_l (2k_l)^{-1} [f_l^+ \exp(ik_l x) + f_l^- \exp(-ik_l x)] \mathbf{e}^l \quad (6')$$

and for  $\mathbf{P} \neq 0$  the solution (4) with one-dimensional  $\mathbf{k}$  and asymptotic value

$$\mathbf{E}(x) \approx 4\pi^2 i \frac{\omega^2}{c^2} \sum_l k_l^{-1} [\theta(x) (\tilde{\mathbf{P}}(k_l) \mathbf{e}^{l*}) \exp(ik_l x) - \theta(-x) (\tilde{\mathbf{P}}(-k_l) \mathbf{e}^{l*}) \exp(-ik_l x)] \mathbf{e}^l. \quad (4''')$$

\* $\text{tg} = \tan$ .

Here  $\theta(\alpha) = 1$  when  $\alpha \geq 0$ , and  $\theta(\alpha) = 0$  when  $\alpha < 0$ .

For an anisotropic medium the analysis becomes somewhat more complicated, owing to the dependence of the phase velocity of the wave on the direction, namely, in anisotropic media the direction  $\nu(\mathbf{n})$  of the energy-flux vector or, which is the same, of the group velocity, does not coincide in general with the direction  $\mathbf{n}$  of the wave vector, and is normal to the surface of the wave vectors (see<sup>[2]</sup>, Sec. 77). Therefore, in a medium with large  $r$  the field at a given point  $\mathbf{r} = r\mathbf{n}'$  is determined by the value of the functions  $f_{\mathbf{l}}(\mathbf{n})$  and  $\tilde{\mathbf{P}}(\mathbf{kn})$  at the same point  $\mathbf{n} = \mathbf{k}/k = \mathbf{n}_a(\mathbf{n}')$ , for which  $\nu(\mathbf{n}_a) = \mathbf{n}'$ . In ordinary crystals, however, the double refraction is small. Moreover, if we are interested in the amplitude of the harmonic in the far field past the exit from the crystal then, in accordance with the considerations advanced in Sec. 2, we must take the Fourier amplitude corresponding precisely to the defined wave vector  $\mathbf{k}$ , and not to the ray vector  $\mathbf{s}$ . The essential differences from the scalar case may occur only near points of internal or external conical refraction. Such a possibility is excluded from consideration here.

Taking the foregoing into account, all the correspondence formulas (3)–(3'), (4)–(4') etc. are applicable here, too. In order to calculate the power carried by the specified solution we must obtain with the aid of this correspondence the asymptotic expression for the fields and the square of the modulus of the amplitude  $\mathbf{E}$  of the outgoing wave must be multiplied by  $(8\pi)^{-1}c\sqrt{\epsilon}r^2d\Omega_{\mathbf{n}}$  for the three-dimensional problem, by  $(8\pi)^{-1}c\sqrt{\epsilon}hrd\varphi$  for the two-dimensional problem, and by  $(8\pi)^{-1}c\sqrt{\epsilon}S$  for the one-dimensional problem. Here  $d\Omega_{\mathbf{n}}$  is the solid-angle element,  $d\varphi$  the linear-angle element,  $h$  the dimension of the cylindrical beam along the cylinder generator, and  $S$  the area of the one-dimensional beam. These formulas are accurate to  $\sim 2\%$  (relative magnitude of the double refraction).

## 2. DESCRIPTION OF SECOND-HARMONIC GENERATION IN THE MOMENTUM REPRESENTATION AND ALLOWANCE FOR THE BOUNDEDNESS OF THE REGION

As is well-known, second-harmonic generation is based on the fact that the laser wave with frequency  $\omega_0$  excites in the so-called "quadratic" medium (see<sup>[1]</sup>) a polarization  $\mathbf{P}$  at frequency  $2\omega_0$ :

$$P_i^{2\omega}(\mathbf{r}) = \chi_{ijkl}^{\omega+\omega} E_k^{\omega}(\mathbf{r}) E_l^{\omega}(\mathbf{r}). \quad (7)$$

Here  $\hat{\chi}$  is the tensor of the quadratic polarizability<sup>5)</sup>.

The transition from the coordinate representation in (7) to the momentum representation calls for some stipulations. Namely, the very separation of the equations for the Fourier components of different momenta  $\mathbf{k}$  in the form (2) presupposes that the medium is infinite. It is nonetheless clear that if the crystal has flat faces and we specify or calculate the field outside the crystal, in the "far field," then all the results of Sec. 1 remain valid here, too.

In this case, however, the exciting force  $\mathbf{P}(\mathbf{r})$ , together with the nonlinear-polarizability tensor, differs from zero only inside the crystal. In this connection it is necessary to expand the spatial distribution  $\hat{\chi}(\mathbf{r})$  itself, which is thus piecewise constant ( $\hat{\chi}(\mathbf{r}) \equiv \hat{\chi}v(\mathbf{r})$ ,  $v(\mathbf{r}) = 1$  inside the crystal and  $v(\mathbf{r}) = 0$  outside), in a three-dimensional integral:

$$\tilde{\hat{\chi}}(\mathbf{q}) \equiv \tilde{\hat{\chi}}v(\mathbf{q}) = \hat{\chi}(2\pi)^{-3} \int e^{-i\mathbf{q}\mathbf{r}} d\mathbf{r}, \quad (8)$$

where the integration is taken over the volume of the crystal. Taking all the foregoing into account, the double-frequency photon with momentum  $\mathbf{k}$  can be obtained from two photons of the laser wave with momenta  $\mathbf{k}_1$  and  $\mathbf{k}'_1$ , acquiring the lacking momentum  $\mathbf{q} = \mathbf{k} - \mathbf{k}_1 - \mathbf{k}'_1$  from the crystal as a whole. By directly calculating the Fourier transform of  $\hat{\chi}v(\mathbf{r})\mathbf{E}(\mathbf{r})\mathbf{E}(\mathbf{r})$ , we obtain

$$\tilde{\mathbf{P}}(\mathbf{k}) = \iint d^3\mathbf{k}_1 d^3\mathbf{k}'_1 \tilde{\hat{\chi}}\tilde{\mathbf{E}}(\mathbf{k}_1)\tilde{\mathbf{E}}(\mathbf{k}'_1)v(\mathbf{k} - \mathbf{k}_1 - \mathbf{k}'_1). \quad (9)$$

If the crystal is bounded by two parallel planes  $x = x_1$  and  $x = x_2$ , through which the radiation enters and leaves, the Fourier transform of such a function  $v(\mathbf{r})$  is

$$\tilde{v}(\mathbf{k}) = \delta(k_y)\delta(k_z)V(k_x), \quad (8a)$$

$$V(q) = (\pi q)^{-1} \exp\left[-iq\frac{(x_1+x_2)}{2}\right] \sin q\frac{x_1-x_2}{2}. \quad (8b)$$

In other words, the plane boundary perpendicular to the  $x$  axis can impart to the photon only an  $x$ -component of additional momentum. Namely, the  $\delta$ -functions in  $\tilde{v}(\mathbf{q})$  from (8a) and in  $\tilde{\mathbf{E}}(\mathbf{k})$  from (3) lower the order of the integration in (9) and

<sup>5)</sup>We note that this tensor, in this (conventional) notation, relates the amplitude of the harmonic components, and if its dispersion would be neglected, it would be twice as large as the tensor of the quadratic polarizability for static fields.

make it possible to obtain the final result. It is easy to see that when  $x_2 - x_1 \rightarrow \infty$  and  $|x_1 + x_2| < \infty$  the function  $\hat{v}(\mathbf{k})$  goes over into  $\delta^3(\mathbf{k})$ ; this corresponds to the fact that in an infinite homogeneous medium the momentum is conserved.

For the one-dimensional problem in a bounded medium the two  $\delta$ -functions in  $\tilde{\mathbf{E}}(\mathbf{k})$  eliminate both integrations, and we obtain the well-known result (see, for example, [1]), even with the correct coefficient (here and below all the formulas are written out for the interaction  $1^0 + 1^0 \rightarrow 2^e$  in a KDP crystal):

$$W_2 = 2^5 \pi^3 n^{-3} c^{-1} (2\omega_0 c^{-1} l)^2 \sin^2 \theta_M \chi_{zyx}^2 \tilde{W}_1^2 S^{-1} \sin^2 \psi / \psi^2, \\ \psi = (k_2 - 2k_1)l/2, \quad l = x_2 - x_1. \quad (10)$$

### 3. TWO-DIMENSIONAL PROBLEM—SECOND-HARMONIC GENERATION IN CYLINDRICALLY FOCUSED BEAMS

Let us consider the two-dimensional problem, eliminating beforehand the coordinate  $z$  and the momentum  $k_z$ . In the case of practical importance, near the synchronism direction, two essentially different methods of orienting the crystal relative to the vertical axis  $z$  are possible.

1. The  $z$  axis of the beam is perpendicular to the line of intersection of the surfaces of the wave vectors. In this case the lengths of the wave vectors of the laser light  $k_1$  and the harmonic  $k_2$  do not depend on the angle  $\varphi$ , and the vertical direction of the beam is chosen such that  $k_2 = 2k_1$ . Such an arrangement is convenient to obtain maximum conversion efficiency.

2. The  $z$  axis is parallel to the line of intersection of the wave-vector surfaces. In this case the magnitude of the wave vector of the harmonic depends on the angle  $\varphi$ ; choosing here the one-dimensional synchronism direction as the reference, we get<sup>6)</sup>

$$k_2(\varphi) \approx 2k_1(1 - \eta\varphi),$$

$\eta$  is the angle of the intersection of the surfaces of the wave vectors. This problem is more interesting for a comparison with a three-dimensional case.

Writing in both cases an integral of type (9) with  $\tilde{v}(\mathbf{q})$  from (8) without  $\delta(k_z)$ , we obtain, integrating all three  $\delta$ -functions

$$\tilde{\mathbf{P}}(\mathbf{k}) \equiv \tilde{\mathbf{P}}(k_2(\varphi), \varphi) = (4k_1)^{-1} \int_{-\pi}^{\pi} \hat{\chi} \mathbf{e} \mathbf{e} (\cos \varphi_1')^{-1} f(\varphi_1) f(\varphi_1') \\ \times V(k_2(\varphi) \cos \varphi - k_1 \cos \varphi_1 - k_1 \cos \varphi_1') d\varphi_1. \quad (11)$$

<sup>6)</sup>In the notation corresponding to the crystal axes,  $\varphi = \theta - \theta_M$ .

In the integral (11)  $\varphi_1'$  is connected with  $\varphi$  and  $\varphi_1$  by the law of conservation of the  $y$ -component of the momentum:

$$k_2(\varphi) \sin \varphi = k_1 \sin \varphi_1 + k_1 \sin \varphi_1'. \quad (12a)$$

If the medium is infinite, then  $V(\mathbf{q}) = \delta(\mathbf{q})$ , and

$$\tilde{\mathbf{P}}(\mathbf{k}) = (2k_1)^{-2} \hat{\chi} \mathbf{e} \mathbf{e} [(\cos \varphi_1')^{-1} + (\cos \varphi_1)^{-1}] \\ \times |\operatorname{tg} \varphi_1 - \operatorname{tg} \varphi_1'|^{-1} f(\varphi_1) f(\varphi_1'), \quad (13)$$

where  $\varphi$ ,  $\varphi_1$ , and  $\varphi_1'$  are connected also with the conservation of the  $x$ -component of the momentum:

$$k_2(\varphi) \cos \varphi = k_1 \cos \varphi_1 + k_1 \cos \varphi_1'. \quad (12b)$$

In (13)  $\varphi_1$  or  $\varphi_1'$  is one of the two solutions of the system (12a)–(12b) (obtainable from each other by substitution  $\varphi_1 \rightleftharpoons \varphi_1'$ ).

For small  $\varphi > 0$  we have in problem 2

$$\varphi_1 = \varphi + \sqrt{2\eta\varphi}, \quad \varphi_1' = \varphi - \sqrt{2\eta\varphi}. \quad (12c)$$

From this it is seen, in particular, that to calculate the intensity of the harmonic in the direction of the one-dimensional synchronism ( $\varphi = 0$ ) in problem 2, and in any direction in problem 1, the concept of an infinite medium is not applicable, since it gives an infinite result.

Starting from general considerations, of the uncertainty-principle type, concerning the connection between the coordinate and momentum representations, we can state that the vector interaction in a focused beam occurs effectively only in a region of several times  $l_{\text{eff}}$  from the focus; here

$$l_{\text{eff}}(\varphi) = (2k_1 - k_2(\varphi))^{-1} \quad (14)$$

(we assume that vector synchronism is allowed, that is,  $k_2(\varphi) < 2k_1$ ). It becomes clear now that the medium can be regarded as infinite only when calculating the intensity of the harmonic in those directions, for which the region indicated above lies completely inside the crystal. On the other hand, for the direction near one-dimensional synchronism  $l_{\text{eff}}$  tends to infinity.

We note that the foregoing reasoning regarding  $l_{\text{eff}}$  is valid to an equal degree also for the three-dimensional problem—spherically focused beams. However, unlike the cylindrical problem, the growth of  $l_{\text{eff}}(\mathbf{n})$ , for  $\mathbf{n}$  close to the direction of one-dimensional synchronism, is not accompanied there by an infinite increase of  $\tilde{\mathbf{P}}(\mathbf{n})$ , and the intensity of the harmonic per unit solid angle is finite for all directions, even assuming that the medium is infinite (see Sec. 4 below).

For a final solution of the problem it is necessary to find in both cases the functions  $f(\varphi)$  from (5). If the laser beam prior to the focusing does

not have too large a divergence<sup>7)</sup> and is focused at an angle  $2\gamma$  much larger than this divergence, then we can use the asymptotic correspondence (5)–(5'), taking for  $u(\mathbf{R}, \varphi)$  simply the field of the laser on leaving the lens. Here and below we shall consider the simplest case of ideal focusing of an ideal beam. Then the function  $f(\varphi)$  (in the three-dimensional case— $f(\mathbf{n})$ ) can be regarded as piecewise constant: it is equal to a constant for those values of  $\varphi$  (or  $\mathbf{n}$ ) in the direction of which there are rays of the laser light, and is equal to zero outside this region; the value of the constant is determined by normalization to the laser-beam power.

We shall not stop to calculate exactly the integral (11) for problem 2 in a bounded medium (for an ideal beam this problem was considered in the coordinate representation in<sup>[4]</sup>). In problem 2, the dependence of the second-harmonic power on the direction near  $\varphi = 0$  is of the form

$$dW_2 = 2^8 \pi^6 n^{-4} (\omega_0/c) c^{-1} \eta^{-1} \sin^2 \theta_M \chi_{zyx}^2 W_1^2 k^{-1} \gamma^{-2} \varphi^{-1} d\varphi. \quad (15)$$

Here  $\varphi$  is the angle of  $\mathbf{k}_2$  inside the crystal; when the harmonic leaves the crystal  $d\varphi_{\text{air}} \approx n(2\omega_0)d\varphi$ .

Formula (15) was obtained for a focus inside an infinite medium, and is therefore valid only when  $\varphi \gtrsim (k_1 l \eta)^{-1}$ ; for smaller values of  $\varphi$  the quantity  $dW_2/d\varphi$  practically stops growing, and when  $\varphi \lesssim -(k_1 l \eta)^{-1}$  it decreases rapidly (in this latter region—with normal dispersion—a nonzero value of  $dW_2/d\varphi$  is obtained only as a result of the contribution of the boundaries of the crystal, and has an oscillating character). For large positive  $\varphi = \varphi_b$  the quantity  $dW_2/d\varphi$  vanishes quite rapidly simply because  $f(\varphi_1) = 0$  for the corresponding  $\varphi_1$ :

$$\varphi_b + \sqrt{2\eta\varphi_b} = \gamma, \quad (16)$$

$\gamma$  is the angular half-width of the beam inside the crystal.

In problem 1, with the beam focused in the center of a crystal of length  $l$ , and under the assumption  $\gamma \gg (kl)^{-1/2}$ , we can readily find that if the directions  $\varphi$  are not too close to the edge of the beam ( $|\varphi - \gamma| \gtrsim (kl)^{-1/2}$ , we get

$$dW_2/d\varphi = 2^7 \pi^5 n^{-3} (\omega_0/c)^2 c^{-1} \sin^2 \theta_M \chi_{zyx}^2 W_1^2 k^{-1} \gamma^{-2} l. \quad (17)$$

#### 4. SPHERICALLY FOCUSED BEAMS: CONICAL LENS

We assume that a laser beam of round cross section is focused so that the direction  $\theta = \theta_M$ ,

$\varphi = 45^\circ$  coincides with the axis of the beam. Then, writing the integral (17) for the case of an infinite medium and eliminating the integration by means of  $\delta$ -functions, we obtain the following result

$$\tilde{\mathbf{P}}(\mathbf{n}) = 2^{-3} (n\omega_0/c)^{-1} \int_0^{2\pi} d\psi \chi_{zyx}^2 e f(\mathbf{n}_1) f(\mathbf{n}_1'). \quad (18)$$

Here

$$\mathbf{n} = \{\theta, \varphi\}, \quad \mathbf{n}_{1,1'} = \{\theta_{1,1'}, \varphi_{1,1'}\} = \{\theta \pm \beta \cos \psi, \varphi \pm \beta \sin \psi\}, \quad \beta \approx \sqrt{2\eta(\theta - \theta_M)}.$$

For (18) to be applicable it is necessary to stipulate, as in Sec. 3, that  $\gamma \gg (kl)^{-1/2}$  and  $l_{\text{eff}}(\mathbf{n}) \ll l$ ; it is easy to see that the first is the condition that the length of the focal region fit well inside the crystal. It is obvious that for  $\theta < \theta_M$  in an infinite medium the intensity of the harmonic is equal to zero, as in the two-dimensional problem. However, in the case when  $\theta \rightarrow \theta_M$ ,  $\theta > \theta_M$  the intensity of the harmonic flattens out, in spite of the growth of  $l_{\text{eff}}(\mathbf{n})$ , and its value is

$$dW_2/d\Omega_2 = 2^9 \pi^7 c^{-1} (\omega_0/c)^2 n^{-3} \chi_{zyx}^2 W_1^2 \Omega_{\pi}^{-2} \sin^2 \theta_M. \quad (19)$$

Here  $\Omega_l = \pi\gamma^2$  is the solid angle of the laser radiation. For a non-ideal beam, the dips on this plateau occur at values of  $\theta$  such that  $2\sqrt{2\eta(\theta - \theta_M)} \sim \beta_k$ . Here  $\beta_k$  is of the order of magnitude of the angular distance at which the phases of the function  $f(\mathbf{n})$  are correlated; that is to say,  $\beta_k$  can be related to the linear distance  $x_k$  of the correlation of the field on the laser front prior to entering a lens with focal distance  $f_{\text{air}}$  (in air):

$$x_k = \beta_{\text{air}} f_{\text{air}} = n\beta_k f_{\text{air}}. \quad (20)$$

On the other hand, for an ideal beam the dips of the plateau occur when the cone of the vector synchronism ( $\mathbf{n}_1 \leftrightarrow \mathbf{n}_1'$ ) goes partially outside the solid angle in which  $f(\mathbf{n})$  differs from zero; for a still larger  $\theta$  no pairs of points ( $\mathbf{n}_1, \mathbf{n}_1'$ ) lying inside the foregoing solid angle remain on the cone at all, and  $dW_2/d\Omega_2 = 0$ . For an ideal laser beam of round cross section, the described regions are shown in the figure; the circle corresponds here to the solid angle of the fundamental radiation, and the doubly or singly shaded regions are respectively of the plateau and of the dip of the harmonic; in this case  $\varphi_b$  is given by (16).

For an approximate calculation of the efficiency of the transformation it is necessary to estimate the order of magnitude of  $\Omega_{2\text{eff}}$ . When  $\gamma \lesssim \eta$  we have

$$\Omega_{2\text{eff}} \approx 0.45 \gamma^3 \eta^{-1} \quad (21)$$

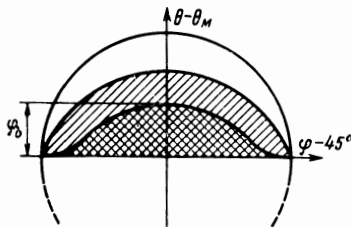
<sup>7)</sup>In the cylindrical model we can already take into account the fact that the beam is not ideal in the horizontal direction.

and then <sup>8)</sup>  $W_2 \sim \gamma^{-1}$ ; at larger values of  $\gamma$ , on the other hand,  $\Omega_{\text{eff}}$  is smaller than given by (21).

As in Sec. 3, the ratio  $dW_2/d\Omega_2$  for  $\theta < \theta_M$  is governed only by the contribution from the crystal boundaries. It is customarily stated that one-dimensional interaction is realized when  $\theta < \theta_M$ ; actually this is not so at all. In particular, the law governing the intensity near the edge of the plateau in the region  $\theta < \theta_M$  has the following form (we take the intensity in the plateau as unity)

$$\frac{dW_2}{d\Omega_2} = \left| \frac{1}{2} - \frac{2}{\pi} \int_0^\psi \frac{\sin x}{x} dx \right|^2.$$

Here  $\psi$  is given by (10); when  $\psi \gtrsim 1$  we have  $dW_2/d\Omega_2 \sim \cos^2 \psi / \psi^2$  in place of  $\sin^2 \psi / \psi^2$ .



Picture showing generation of second harmonic in a spherically focused ideal beam in an infinite medium. Circle – direction of laser radiation. Doubly and singly shaded areas correspond respectively to the plateau and the dip of the harmonic; the value of  $\psi_b$  is taken from (16).

We note that in the three-dimensional problem the growth of  $l_{\text{eff}}$  leads to a growth of  $dW_2/d\Omega_2$  only for focusing near one of the boundaries of the crystal instead of in its center. In this case expression (19) must be multiplied by a quantity of the order of  $(1 + |\ln(a_1/a_2)|)^2$ , where  $a_1$  and  $a_2$  are the distance to the focus from the first and second boundaries. Naturally, such a distance cannot be regarded as smaller than the length of the focal spot  $(k\gamma^2)^{-1}$ , so that the maximum gain due to focusing on the boundary is of the order of  $(1 + \ln kl + 2 \ln \gamma)^2$  and is small in a real situation. Obviously, with such a focusing the distribution of the harmonic becomes smeared out over angles  $\Delta\alpha \sim (ka_{\text{min}}\eta)^{-1}$  compared with (18).

We have previously proposed<sup>[5]</sup> to use a conical lens to realize vector synchronism, and we calculated there in the coordinate representation the

laser field in such a system. The momenta of the photons focused by a conical lens lie almost all on the same vector-synchronism cone. Estimating for an ideal laser beam of diameter  $D$

$$\Omega_l \sim (kD/2)2\pi \sin \beta_1, \quad \Omega_2 \approx \pi(kD/2)^{-2},$$

we obtain, in accord with (18)–(19)

$$W_2/W_1 \approx 2^7 \pi^6 c^{-4} n^{-3} (\omega_0/c)^2 \chi_{zyx}^2 W_1 (\sin \beta_1)^{-2}. \quad (22)$$

We have chosen here the axis of the conical lens in the direction  $\theta = 90^\circ$ ,  $\varphi = 45^\circ$ .

With such a generation method, the light of the emerging harmonic has a very small angular width; for an ideal input beam it is of the order of its ideal width. The necessary angle  $\beta_1$  for the convergence of the rays to the beam axis in the crystal can be determined from the equation

$$k_{2\omega}^e = 2k_{\omega}^o \cos \beta_1; \quad (23)$$

The necessary length of the crystal  $l \approx D/2 \tan \beta_1$  under ordinary conditions (KDP crystal, ruby laser,  $D \sim 1$  cm) does not exceed 3 cm, but the orientation of the plane faces must be perpendicular to the direction indicated above. Numerical estimates by means of formula (22) yield for the ideal beam

$$W_2/W_1 = W_1/A, \quad A \approx (\chi_{zyx}/10^{-9} \text{CGSE})^{-2} \cdot 1.6 \cdot 10^5 W.$$

## 5. EFFICIENCY OF SECOND-HARMONIC GENERATION BY AN IDEAL BEAM

We have solved several problems involving the second-harmonic generation for different methods of laser-beam focusing. This raises the natural question of finding the optimal generation method. To this end we calculate in this section, for an ideal laser beam in the specified-field approximation, the efficiency of transformation as a function of the system parameters: crystal length, beam diameter, focusing angle, etc. The maximum efficiency for each method of generation corresponds to limiting values of the parameters; near these values, the formulas themselves cease to be exact. In this connection, the results of this section are tentative in character.

1) One-dimensional interaction. We must first indicate the limit of applicability of the strictly one-dimensional model of interaction of the laser wave and the harmonic. We assume the laser beam to be ideally parallel, that is, its angle of divergence is  $\alpha \approx \lambda/d$  ( $d$  is the transverse dimension of the beam). We stipulate in this connection that the divergence of the beam  $\alpha$  not lead to an appreciable change in the transverse dimension  $d$  of the laser wave and, by the same token, of the field amplitude in it over the crystal length  $l$ :

<sup>8)</sup>It may turn out in the experiment that the region of applicability of formula (21) corresponds to angles  $\gamma$  much smaller than those at which the formulas (18)–(19) are by themselves valid (owing to the boundedness of the crystal and the nonideal nature of the beam).

$$al \lesssim d \rightarrow d \gtrsim D_1 = \sqrt{l\lambda}.$$

We also stipulate that no "aperture effects" be noticeable (see<sup>[6]</sup>), since an analysis of these effects necessitates going beyond the framework of the one-dimensional model. To this end it is necessary that the shift of the harmonic beam relative to the laser beam, over the length of the crystal, not exceed the "vertical" transverse dimension  $d_2$  of the laser beam:

$$d_2 \gtrsim D_2 = \eta l.$$

We note that at lengths  $l \gtrsim L = \lambda_0 n^{-1} \eta^{-2}$  (which amounts in KDP to  $\sim 1$  mm)  $D_1 < D_2$ , that is, the second requirement is more restrictive.

As a consequence of these requirements, we find that the quantity  $\sin \psi / \psi$ , which enters in the formulas of the one-dimensional problem, is the same for all the three-dimensional Fourier components that constitute the one-dimensional beam. We can therefore put  $\psi = 0$ ,  $\sin \psi / \psi = 1$ . The beam area is  $S = \pi d_1 d_2 / 4$ . It is advantageous to consider here the following two methods.

A. If we use ordinary telescopic systems, then a beam of round cross section can be contracted to a limiting diameter  $d_1 = d_2 = D_2$ , and then we have from (10)

$$W_2 / W_1 = B \cdot 0.4 \eta^{-2} \sin^2 \theta_M, \quad (24)$$

$$B = 2^7 \pi^4 c^{-1} n^{-3} (\omega_0 / c)^2 \chi_{zyx}^2 W_1.$$

B. Since the aperture effects operate in the vertical direction, we can visualize a telescopic system that contracts the beam to  $d_1 = D_1$  in one direction and to  $d_2 = D_2$  in the other; then

$$W_2 / W_1 = B \cdot 0.4 \eta^{-1} (\ln \omega_0 / c)^{1/2} \sin^2 \theta_M. \quad (25)$$

2) Cylindrically focused beams. We consider the orientation method 1 of Sec. 3. In deriving formula (17) it has been assumed that there are no aperture effects in the vertical direction ( $h \gtrsim D_2$ ) and that the length of the focal region when the beam is focused at an angle  $\gamma$  lies entirely inside the crystal:  $(n \omega_0 c^{-1} \gamma^2)^{-1} \lesssim L$ , which is equivalent to the condition  $d_1 = \gamma l \gtrsim \sqrt{l\lambda} / 2\pi$ . In other words, we obtain method B of case 1, but now as the limiting case of cylindrical focusing. Substituting the minimum values of  $h$  and  $\gamma$ , we obtain a result with a coefficient that differs somewhat from (25):

$$W_2 / W_1 = B \cdot 3.14 \eta^{-1} (n \omega_0 l / c)^{1/2} \sin^2 \theta_M. \quad (26)$$

3) Spherically focused beams. For applicability of formula (19), even in half the solid angle  $\Omega_{2 \text{ eff}}$ , it is necessary to have

$$l_{\text{eff}} = (2k_1 - k_2(\mathbf{n}))^{-1} = (2\eta k_1 \varphi_b)^{-1} = (n \omega_0 \gamma^2 c^{-1})^{-1} \lesssim l.$$

Further, if  $l \gtrsim L$  and  $\gamma \lesssim \eta$ , then  $\Omega_{2 \text{ eff}}$  is determined from (21). Substituting the minimum value of  $\gamma$ , we obtain

$$W_2 / W_1 = B \cdot 5.7 \eta^{-1} (n \omega_0 l / c)^{1/2} \sin^2 \theta_M. \quad (27)$$

We note that although (27) differs from (26) and (25) only by a small factor, the generation picture differs here greatly from that described in case 2 and in case 1 (method B). Namely, here  $d_1 = d_2 = D_1$ . The harmonic beam then shifts strongly relative to the laser beam; formulas (19) and (21) take these aperture effects into account automatically.

4) Conical lens. For a conical lens (see<sup>[5]</sup>) the efficiency and the limitations were obtained in Sec. 4. We present them here:

$$W_2 / W_1 = B \cdot 10 (\sin \beta_1)^{-2}, \quad (28)$$

and in this case

$$D \lesssim 2l \operatorname{tg} \beta_1, \quad n^e(2\omega_0) = n^o(\omega_0) \cos \beta_1,$$

in a KDP crystal  $\beta_1 \approx 0.12$ .

We present numerical values for the coefficients of  $B$  in formulas (24)–(28) for a KDP crystal of length  $l = 3$  cm: these are  $2 \times 10^2$ ,  $3.5 \times 10^3$ ,  $2.8 \times 10^4$ ,  $5 \times 10^4$ , and  $6 \times 10^2$  in (24), (25), (26), (27) and (28), respectively. We see that the best efficiency of transformation is obtained with spherical focusing; somewhat worse values are obtained with cylindrical focusing and telescoping. We note, however, that the dependence on  $l$  and  $\eta$  for these three cases is the same. One cannot exclude that such a strong difference in the coefficients of  $\eta^{-1} l^{1/2}$  is connected not only with the effective advantage of one generation method over the other, but also with the uncertainty in the estimate of the limiting parameters (of the type  $\alpha = \lambda/D$  or  $\alpha = \lambda/D = \lambda/2\pi D$ ).

For the methods described for cases 1–3, the beam should be contracted quite strongly:  $D_1 = \eta l \approx 0.1$  cm,  $D_2 = \sqrt{l\lambda} \approx 0.1$  cm, and the transformation efficiency decreases rapidly with increasing parameters, compared with the limiting values. At the same time, for a conical lens  $D = 2l \tan \beta_1 \approx 0.8$  cm and the efficiency does not depend on  $D$  at all. We note also that, owing to the inherent divergence of the laser beam, the usual focusing and telescoping may not give the desired effect, while for the conical lens the situation is somewhat more favorable. In this connection the generation of the second harmonic with the aid of the conical lens is quite promising.

Almost everywhere above we assume the laser beam to be ideal. For a nonideal beam, on the

other hand, it is necessary to have some sort of model of the function  $f(\mathbf{n})$ ; at the present time we have no such model (see the discussion in<sup>[5]</sup>).

Essentially, the function  $f(\mathbf{n})$  should be regarded as a random function. Knowledge of its autocorrelation function

$$R(\mathbf{n}_1, \mathbf{n}_2) = \overline{f(\mathbf{n}_1)f^*(\mathbf{n}_2)}$$

makes it possible to represent  $f(\mathbf{n})$  in the form of a series in fixed functions  $g_j(\mathbf{n})$  with noncorrelated coefficients. As is well known, the functions  $g_j(\mathbf{n})$  are solutions of the integral equation

$$\int R(\mathbf{n}_1, \mathbf{n}_2)g(\mathbf{n}_2)d\mathbf{n}_2 = \Lambda g(\mathbf{n}_1).$$

Thus, to study a laser beam it is quite advantageous to determine experimentally the function  $R(\mathbf{n}_1, \mathbf{n}_2)$ .

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<sup>1</sup>S. A. Akhmanov and R. V. Khokhlov, *Problemy nelineinoi optiki* (Problems of Nonlinear Optics), INI AN SSSR, 1964.

<sup>2</sup>L. D. Landau and E. M. Lifshitz, *Élektrodinamika sploshnykh sred* (Electrodynamics of Continuous Media), Gostekhizdat, 1957.

<sup>3</sup>L. D. Landau and E. M. Lifshitz, *Kvantovaya mekhanika* (Quantum Mechanics), Fizmatgiz, 1964.

<sup>4</sup>S. A. Akhmanov, A. P. Sukhorukov, and R. V. Khokhlov, *JETP* **50**, 474 (1966), *Soviet Phys. JETP* **23**, 316 (1966).

<sup>5</sup>B. Ya. Zel'dovich, N. F. Pilipetskiĭ, *Izv. Vuzov Radiofizika* **9**, No. 1, 1966.

<sup>6</sup>D. G. Boyd, A. Ashkin, J. M. Dziedzic, and D. A. Kleinman, *Phys. Rev.* **137**, A1305 (1965).

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