

Optical-Mode Interaction in Nonlinear Media*

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(Received 8 May 1969)

A general method of approach to nonlinear optical problems involving traveling-wave beams produced by the spherical Fabry-Perot-type open resonator has been developed. It is shown how coupled-mode equations can be obtained for the nonlinear interaction of these modes from Maxwell's equations describing the wave propagation in the nonlinear medium. The coupling coefficients for second-harmonic generation are calculated as an example of the use of the method. From these general results, effects arising from position and strength of focusing may be obtained.

1. INTRODUCTION

PREVIOUSLY¹⁻⁴ the theory of nonlinear optical effects has been formulated in terms of plane-wave interactions. The effects produced by focused beams have been studied by analyzing the beam into its constituent plane waves and then appealing to the plane-wave theory.⁵⁻⁸ This method has been used with success by Boyd and Kleinman⁹ (BK) to study second-harmonic generation in the small-conversion approximation in detail. Since the method precludes studying the interaction between two or more focused beams, it cannot be used in a more complicated situation. For example, BK have to make a drastic approximation which is valid only for the very small gain region when applying the method to parametric amplification when there is interaction between the signal and idler beams. The basic difficulty arises since the plane-wave method leads to an infinite set of coupled differential equations for the amplitudes of the constituent plane waves in each beam. This set of equations cannot be approximated in any sensible fashion.

We put forward in this paper a method by which this problem may be overcome. We show that by basing the analysis on the modes of the open optical resonator, which are the natural modes of the physical situation, a set of differential equations may be obtained which can be approximated in a logical fashion. Boyd and Gordon¹⁰ and Boyd and Kogelnik¹¹ first derived the

Hermite-function approximation to these modes by a diffraction-theory approach, and later Kogelnik and Li¹² (KL) derived the same form from the scalar wave equation. KL considered an almost plane wave propagating in a homogeneous isotropic linear dielectric in the positive z direction. In the absence of absorption, the electric field of this wave may then be written in the form

$$E(x, y, z) = \mathcal{E}(x, y, z)e^{-ikz}, \quad (1.1)$$

where $k = 2\pi/\lambda$, and $\mathcal{E}(x, y, z)$ is a slowly varying function of x , y , and z which represents the differences between the waveform considered and a plane wave. Substituting this form into the Helmholtz equation arising from the wave equation and making the approximation

$$|\partial^2 \mathcal{E} / \partial z^2| \ll k |\partial \mathcal{E} / \partial z|, \quad (1.2)$$

they obtained the following equation for $\mathcal{E}(x, y, z)$:

$$\frac{\partial^2 \mathcal{E}}{\partial x^2} + \frac{\partial^2 \mathcal{E}}{\partial y^2} - 2ik \frac{\partial \mathcal{E}}{\partial z} = 0. \quad (1.3)$$

KL obtained a set of solutions to this equation which may be written in the form

$$\mathcal{E}_{nm} = \frac{\sqrt{2}(1+i\xi)^{(n+m)/2}}{w_0(2^{n+m}n!m!\pi)^{1/2}(1-i\xi)^{(n+m)/2+1}} \times H_n\left(\frac{x\sqrt{2}}{w}\right) H_m\left(\frac{y\sqrt{2}}{w}\right) \exp\left(\frac{-(x^2+y^2)}{w_0^2(1-i\xi)}\right), \quad (1.4)$$

where $\xi = 2(z-f)/kw_0^2$, $w = w_0(1+\xi^2)^{1/2}$, $z=f$ denotes the focus position, w_0 the spot size at the focus position, and $H_n(x)$ the Hermite polynomial of degree n . Multiplied by the factor e^{-ikz} , this equation then describes the form of the traveling wave derived from the nm th mode of a resonator cavity which is centered on the axis $x=0$, $y=0$. The traveling wave may be considered as part of the standing wave in the resonator cavity, or the wave which propagates into space when one of the mirrors of the resonator is partially transmitting.

* Research supported in part by the Ministry of Technology (Great Britain) and by the U. S. Army Research Office (Durham).

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¹ N. Bloembergen, *Non-Linear Optics* (W. A. Benjamin, Inc., New York, 1965).

² J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. Pershan, *Phys. Rev.* **127**, 1918 (1962).

³ D. A. Kleinman, *Phys. Rev.* **128**, 1761 (1962).

⁴ P. N. Butcher, *Nonlinear Optical Phenomena*, Bulletin 200, Engineering Experiment Station, Ohio State University, Columbus, Ohio (unpublished).

⁵ D. A. Kleinman and G. D. Boyd, *Phys. Rev.* **145**, 338 (1966).

⁶ D. A. Kleinman and R. C. Miller, *Phys. Rev.* **148**, 302 (1966).

⁷ G. D. Boyd, A. Ashkin, J. M. Dziedzic, and D. A. Kleinman, *Phys. Rev.* **137**, 1305 (1962).

⁸ G. D. Boyd and A. Ashkin, *Phys. Rev.* **146**, 187 (1966).

⁹ G. D. Boyd and D. A. Kleinman, *J. Appl. Phys.* **39**, 3597 (1968).

¹⁰ G. D. Boyd and J. P. Gordon, *Bell System Tech. J.* **40**, 489 (1961).

¹¹ G. D. Boyd and H. Kogelnik, *Bell System Tech. J.* **41**, 1347 (1962).

¹² H. Kogelnik and T. Li, *Appl. Opt.* **5**, 1550 (1966).

Using the orthonormality relations of the Hermite functions, we may derive the relations

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{E}_{nm}^* \mathcal{E}_{rs} dx dy = \delta_{nr} \delta_{ms}. \quad (1.5)$$

Subsequently, the analysis is restricted to the class of traveling-wave fields which may be written as a sum of the resonator modes:

$$E = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} A_{nm} \mathcal{E}_{nm}(x, y, z) e^{-ikz}. \quad (1.6)$$

Thus for the optical fields considered these traveling-wave modes form a complete orthonormal set. For the fields of interest, those inside an optical resonator and those emanating from an optical resonator through a partially transmitting end mirror, an expansion of the form (1.6) is obviously possible.

2. PROPAGATION IN UNIAXIAL MEDIUM

The method is developed for nonlinear interactions in uniaxial media, since this is the most important case, but the general approach is obviously valid for isotropic media where the equations are simpler. We consider a beam propagating in the positive z direction in a uniaxial medium which is oriented so that its optic axis is perpendicular to the axis of the beam and is in the x direction. It is assumed that phase matching may be achieved with this configuration. Lithium niobate is taken as an example of a material for which this criterion is satisfied. The method may be generalized to the case when the beams propagate at some angle to the optic axis, but then the assumption of small anisotropy has to be made.

The axes chosen are principal axes of the dielectric tensor, which therefore may be written

$$\epsilon = \begin{pmatrix} \epsilon_x & 0 & 0 \\ 0 & \epsilon_z & 0 \\ 0 & 0 & \epsilon_z \end{pmatrix}. \quad (2.1)$$

Maxwell's equations governing the propagation in a linear medium at frequency ω may be written

$$\nabla \times (\nabla \times \mathbf{E}) - (\omega^2/c^2) \epsilon \cdot \mathbf{E} = 0, \quad (2.2)$$

and the divergence equation is

$$\nabla \cdot (\epsilon \cdot \mathbf{E}) = 0. \quad (2.3)$$

In the two-dimensional problem when there is no variation of the fields in the y direction, i.e., $\partial/\partial y \equiv 0$, these equations split into two groups. The first for the ordinary wave is

$$\frac{\partial^2 E_y}{\partial x^2} + \frac{\partial^2 E_y}{\partial z^2} - \frac{\omega^2}{c^2} \epsilon_z E_y = 0, \quad (2.4)$$

and the second for the extraordinary wave is

$$\frac{\partial^2 E_z}{\partial x \partial z} - \frac{\partial^2 E_x}{\partial z^2} - \frac{\omega^2}{c^2} \epsilon_x E_x = 0, \quad (2.5a)$$

$$\frac{\partial^2 E_x}{\partial x \partial z} - \frac{\partial^2 E_z}{\partial x^2} - \frac{\omega^2}{c^2} \epsilon_z E_z = 0, \quad (2.5b)$$

$$\epsilon_x \frac{\partial E_x}{\partial x} + \epsilon_z \frac{\partial E_z}{\partial z} = 0. \quad (2.5c)$$

Following the method of KL, each set of equations may be reduced to a two-dimensional equation of the form of Eq. (1.3) for a set of resonator modes. The derivation for the ordinary set follows exactly that of KL. For the extraordinary set (2.5), we put

$$\begin{aligned} E_x(x, y, z) &= \mathcal{E}_x(x, y, z) e^{-ik_e z}, \\ E_z(x, y, z) &= \mathcal{E}_z(x, y, z) e^{-ik_e z}, \end{aligned} \quad (2.6)$$

where $k_e^2 = (\omega^2/c^2) \epsilon_x$. From (2.5c) we see that

$$|E_z| \sim k^{-1} |E_x|, \quad (2.7)$$

and hence the longitudinal field may be neglected, as in the three-dimensional isotropic case. Substituting from (2.6) and (2.5c) into (2.5a) and making the approximation (1.2), we obtain the equation

$$\frac{\epsilon_x}{\epsilon_z} \frac{\partial^2 \mathcal{E}_x}{\partial x^2} - 2ik_e \frac{\partial \mathcal{E}_x}{\partial z} = 0. \quad (2.8)$$

This equation leads to a set of modes in the usual way. These modes have been studied in detail by Bhawalkar, Goncharenko, and Smith.¹³ Thus a two-dimensional resonator supports two independent sets of modes with orthogonal linear polarization. The spot sizes of the two sets are related by

$$(w_0)_{\text{extraord}} = \left(\frac{\epsilon_x}{\epsilon_z} \right)^{1/2} (w_0)_{\text{ord}}. \quad (2.9)$$

In three dimensions the situation is more complicated since the extraordinary and ordinary waves do not propagate independently. However, it is possible to define a set of ordinary modes in the absence of an extraordinary polarized field and vice versa. Equations (2.2) and (2.3) written in full are

$$\frac{\partial^2 E_x}{\partial x \partial z} + \frac{\partial^2 E_y}{\partial x \partial y} - \frac{\partial^2 E_x}{\partial z^2} - \frac{\partial^2 E_x}{\partial y^2} - \frac{\omega^2}{c^2} \epsilon_x E_x = 0, \quad (2.10a)$$

$$\frac{\partial^2 E_x}{\partial y \partial x} + \frac{\partial^2 E_z}{\partial z \partial y} - \frac{\partial^2 E_y}{\partial x^2} - \frac{\partial^2 E_y}{\partial z^2} - \frac{\omega^2}{c^2} \epsilon_z E_y = 0, \quad (2.10b)$$

¹³ D. D. Bhawalkar, A. M. Goncharenko, and R. C. Smith, Brit. J. Appl. Phys. 18, 1431 (1967).

$$\frac{\partial^2 E_y}{\partial z \partial y} + \frac{\partial^2 E_x}{\partial z \partial x} - \frac{\partial^2 E_z}{\partial y^2} - \frac{\partial^2 E_z}{\partial x^2} - \frac{\omega^2}{c^2} \epsilon_z E_z = 0, \quad (2.10c)$$

and

$$\epsilon_x \frac{\partial E_x}{\partial x} + \epsilon_z \frac{\partial E_y}{\partial y} + \epsilon_z \frac{\partial E_z}{\partial z} = 0. \quad (2.10d)$$

It may be seen immediately that there exists a solution for which the extraordinary field is identically zero. Then the equations reduce to

$$\frac{\partial}{\partial x} \left[\frac{\partial E_y}{\partial y} + \frac{\partial E_z}{\partial z} \right] = 0, \quad (2.11a)$$

$$\frac{\partial^2 E_z}{\partial z \partial y} - \frac{\partial^2 E_y}{\partial x^2} - \frac{\partial^2 E_y}{\partial z^2} - \frac{\omega^2}{c^2} \epsilon_z E_y = 0, \quad (2.11b)$$

$$\frac{\partial^2 E_y}{\partial z \partial x} - \frac{\partial^2 E_z}{\partial y^2} - \frac{\partial^2 E_z}{\partial x^2} - \frac{\omega^2}{c^2} \epsilon_z E_z = 0, \quad (2.11c)$$

$$\frac{\partial E_y}{\partial y} + \frac{\partial E_z}{\partial z} = 0. \quad (2.11d)$$

Equations (2.11b)–(2.11d) are just the equations which arise from Eq. (2.2) in the case of an isotropic medium. As before, by making the substitution

$$E_{y,z} = \mathcal{E}_{y,z} e^{-ik_0 z}, \quad (2.12)$$

where $k_0^2 = (\omega^2/c^2) \epsilon_z$, and making the approximation (1.2), we obtain

$$\frac{\partial^2 \mathcal{E}_y}{\partial x^2} + \frac{\partial^2 \mathcal{E}_y}{\partial y^2} - 2ik_0 \frac{\partial \mathcal{E}_y}{\partial z} = 0. \quad (2.13)$$

From (2.11d), $|E_z| \sim k_0^{-1} |E_y|$, and the longitudinal field may be neglected. There does not exist a solution of Eq. (2.10) for which the ordinary field is identically zero; however, there is a solution consistent with the assumptions

$$E_{x,y,z} = \mathcal{E}_{x,y,z} e^{-ik_0 z} \quad (2.14a)$$

and

$$|E_y| \sim |E_z| \sim k^{-1} |E_x|. \quad (2.14b)$$

Neglecting terms of order $k^{-1} |E_x|$ in Eqs. (2.10) leads to

$$\frac{\partial^2 E_z}{\partial x \partial z} - \frac{\partial^2 E_x}{\partial z^2} - \frac{\partial^2 E_x}{\partial y^2} - \frac{\omega^2}{c^2} \epsilon_x E_x = 0, \quad (2.15a)$$

$$\frac{\partial^2 E_x}{\partial y \partial x} + \frac{\partial^2 E_z}{\partial y \partial z} - \frac{\partial^2 E_y}{\partial z^2} - \frac{\omega^2}{c^2} \epsilon_z E_y = 0, \quad (2.15b)$$

$$\frac{\partial^2 E_y}{\partial z \partial y} + \frac{\partial^2 E_x}{\partial z \partial x} - \frac{\omega^2}{c^2} \epsilon_z E_z = 0, \quad (2.15c)$$

$$\epsilon_x \frac{\partial E_x}{\partial x} + \epsilon_z \frac{\partial E_z}{\partial z} = 0. \quad (2.15d)$$

Substituting from (2.15d) and neglecting Eq. (2.15c) for the longitudinal component, we obtain

$$\frac{\epsilon_x}{\epsilon_z} \frac{\partial^2 E_x}{\partial x^2} + \frac{\partial^2 E_x}{\partial y^2} + \frac{\partial^2 E_x}{\partial z^2} + \frac{\omega^2}{c^2} \epsilon_x E_x = 0 \quad (2.16a)$$

and

$$\frac{\partial^2 E_y}{\partial z^2} + \frac{\omega^2}{c^2} \epsilon_z E_y = \left(1 - \frac{\epsilon_x}{\epsilon_z} \right) \frac{\partial^2 E_x}{\partial x \partial y}. \quad (2.16b)$$

Equation (2.16a) leads to a set of modes in the usual way. Note that because of the ϵ_x/ϵ_z factor the surfaces of constant phase will be ellipsoidal and not spherical as in the usual case. Thus a resonator designed to support the extraordinary modes must have ellipsoidal mirrors. This was first pointed out by Bhawalkar *et al.*¹³

Substituting Eq. (2.14) into (2.16a) gives

$$-2ik_0 \frac{\partial \mathcal{E}_y}{\partial z} = \left(1 - \frac{\epsilon_x}{\epsilon_z} \right) \frac{\partial^2 \mathcal{E}_x}{\partial x \partial y} e^{-i(k_0 - k_0)z}. \quad (2.17)$$

Since $k_0 \neq k_o$, the magnitude of the ordinary wave cannot grow but just oscillates throughout the medium and remains negligible.

Because the two different sets of modes are supported by differently shaped end mirrors to the cavity, a given cavity can only support one set polarized either as an ordinary or an extraordinary wave.

3. COUPLED-MODE EQUATIONS

In a nonlinear-medium equation (2.1) becomes

$$\nabla \times (\nabla \times \mathbf{E}) - (\omega^2/c^2) \mathbf{E} = (4\pi\omega^2/c^2) \mathbf{P}^{NL}, \quad (3.1)$$

where \mathbf{P}^{NL} denotes the polarization arising from the nonlinear response of the dielectric at frequency ω . Under the same condition the divergence equation becomes

$$\nabla \cdot (\mathbf{E} \cdot \mathbf{E}) = -4\pi \nabla \cdot \mathbf{P}^{NL}. \quad (3.2)$$

Proceeding exactly as in the linear case, we obtain the equations

$$\frac{\partial^2 \mathcal{E}_y}{\partial x^2} + \frac{\partial^2 \mathcal{E}_y}{\partial y^2} - 2ik_0 \frac{\partial \mathcal{E}_y}{\partial z} = -\frac{4\pi\omega^2}{c^2} P_y^{NL} e^{ik_0 z} \quad (3.3a)$$

and

$$\frac{\epsilon_x}{\epsilon_z} \frac{\partial^2 \mathcal{E}_x}{\partial x^2} + \frac{\partial^2 \mathcal{E}_x}{\partial y^2} - 2ik_0 \frac{\partial \mathcal{E}_x}{\partial z} = -\frac{4\pi\omega^2}{c^2} P_x^{NL} e^{ik_0 z} \quad (3.3b)$$

for the ordinary and extraordinary beams, respectively. The term arising from $\nabla \cdot \mathbf{P}^{NL}$ has been neglected, since \mathbf{P}^{NL} will, in general, arise from fields which vary slowly in the x - y direction, and the relations

$$\left| \frac{\partial}{\partial x} \nabla \cdot \mathbf{P}^{NL} \right| \ll \frac{4\pi\omega^2}{c^2} |P_x^{NL}|, \quad (3.4)$$

$$\left| \frac{\partial}{\partial y} \nabla \cdot \mathbf{P}^{NL} \right| \ll \frac{4\pi\omega^2}{c^2} |P_y^{NL}|$$

will hold. Since the nonlinear polarization is a small perturbation in Eqs. (3.3), the coupled-mode approach¹⁴ may be used for their solution. Thus writing the fields

$$\mathcal{E}_y = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} A_{nm}(z) \mathcal{E}_{nm}^o(x, y, z), \quad (3.5a)$$

$$\mathcal{E}_x = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} B_{nm}(z) \mathcal{E}_{nm}^e(x, y, z), \quad (3.5b)$$

where the coefficients A_{nm} and B_{nm} are slowly varying functions of z , leads to the equations

$$\sum_{n=0}^{\infty} \sum_{m=0}^{\infty} -2ik_o \frac{dA_{nm}}{dz} \mathcal{E}_{nm}^o = -\frac{4\pi\omega^2}{c^2} P_y^{NL} e^{ik_o z}, \quad (3.6a)$$

$$\sum_{n=0}^{\infty} \sum_{m=0}^{\infty} -2ik_e \frac{dB_{nm}}{dz} \mathcal{E}_{nm}^e = -\frac{4\pi\omega^2}{c^2} P_x^{NL} e^{ik_e z}, \quad (3.6b)$$

where the second derivatives of the coefficients have been neglected.

Using the orthonormality of the modes, we obtain the equations

$$\frac{dA_{nm}}{dz} = -\frac{2\pi i \omega^2}{k_o c^2} e^{ik_o z} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{E}_{nm}^{o*} P_y^{NL} dx dy, \quad (3.7a)$$

$$\frac{dB_{nm}}{dz} = -\frac{2\pi i \omega^2}{k_e c^2} e^{ik_e z} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{E}_{nm}^{e*} P_x^{NL} dx dy \quad (3.7b)$$

for the rate of change of the amplitude of each mode of the field due to the nonlinear polarization. These equations apply to any nonlinear interaction. To proceed further, we must specify the functional form of the nonlinear polarization in order to evaluate the integrals. We will consider second-harmonic generation (SHG) as the simplest case and then briefly parametric amplification.

4. SECOND-HARMONIC GENERATION

Second-harmonic generation arises from the polarization quadratic in the electric field. Taking lithium niobate as our example, the relevant coupling polarization is given by¹⁵

$$P_y^{\omega} = d_{15} E_y^{\omega*} E_x^{2\omega}, \quad (4.1a)$$

$$P_x^{2\omega} = \frac{1}{2} d_{31} E_y^{\omega} E_y^{\omega*}. \quad (4.1b)$$

The ordinary field at the fundamental frequency ω is coupled to the extraordinary field at the harmonic frequency 2ω , and other couplings are negligible because of the phase-matching effect.

¹⁴ W. H. Louisell, *Coupled Mode Theory and Parametric Electronics* (John Wiley & Sons, Inc., New York, 1960).

¹⁵ G. D. Boyd, R. C. Miller, K. Nassau, W. L. Bond, and A. Savage, *Appl. Phys. Letters* **5**, 234 (1964).

Substituting from Eqs. (1.6) into (4.1), we obtain the form for the nonlinear polarization

$$P_y^{\omega} = d_{15} \sum_{j,k,r,s} A_{rs}^{\omega*} B_{kl}^{2\omega} \mathcal{E}_{rs}^{\omega*} \mathcal{E}_{kl}^{2\omega} e^{-i(k_2 - k_1)z}, \quad (4.2a)$$

$$P_x^{2\omega} = \frac{1}{2} d_{31} \sum_{j,k,r,s} A_{rs}^{\omega} A_{kl}^{\omega} \mathcal{E}_{rs}^{\omega} \mathcal{E}_{kl}^{\omega} e^{-i2k_1 z}, \quad (4.2b)$$

where k_2 is the wave number of the extraordinary beam at frequency 2ω , and k_1 is the wave number of the ordinary beam at the fundamental frequency ω .

These expressions may now be substituted into Eq. (3.7) to obtain the coupled-mode equations for SHG:

$$dA_{mn}^{\omega}/dz = C_{mnrskl}^{\omega} A_{rs}^{\omega*} B_{kl}^{2\omega}, \quad (4.3a)$$

$$dB_{mn}^{2\omega}/dz = C_{mnrskl}^{2\omega} A_{rs}^{\omega} A_{kl}^{\omega}, \quad (4.3b)$$

where we have used the summation convention, and the coupling coefficients are defined by the equations

$$C_{mnrskl}^{\omega} = -\frac{2\pi i \omega^2 d_{15}}{k_1 c^2} e^{-i\Delta k z} \times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{E}_{mn}^{\omega*} \mathcal{E}_{rs}^{\omega*} \mathcal{E}_{kl}^{2\omega} dx dy, \quad (4.4a)$$

$$C_{mnrskl}^{2\omega} = -\frac{4\pi i \omega^2 d_{31}}{k_2 c^2} e^{i\Delta k z} \times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{E}_{mn}^{2\omega*} \mathcal{E}_{rs}^{\omega} \mathcal{E}_{kl}^{\omega} dx dy, \quad (4.4b)$$

where $\Delta k = k_2 - 2k_1$.

The form of the coupling coefficients suggests the relation

$$\eta_{\omega} C_{mnrskl}^{\omega} = -\eta_{2\omega} C_{mnrskl}^{2\omega*} \quad (4.5)$$

for interaction in a lossless medium when Kleinman's symmetry condition relates $d_{31} = d_{15}$. This relation may be proved, in general, as follows.

The total energy flux carried by the two fields is represented by the expression

$$\sum_{n=0}^{\infty} \sum_{m=0}^{\infty} (\eta_{\omega} A_{mn}^{\omega} A_{mn}^{\omega*} + \eta_{2\omega} B_{mn}^{2\omega} B_{mn}^{2\omega*}) \quad (4.6)$$

(where η denotes the refractive index), which must remain independent of z in a lossless medium. Differentiating this expression and substituting from Eq. (4.3), we obtain, on rearranging the dummy indices, the equation

$$(\eta_{\omega} C_{mnrskl}^{\omega} + \eta_{2\omega} C_{mnrskl}^{2\omega*}) A_{mn}^{\omega*} A_{rs}^{\omega*} B_{kl}^{2\omega} + \text{c.c.} = 0. \quad (4.7)$$

This relation must hold for all possible values of the mode amplitudes; hence Eqs. (4.5) must be satisfied by the coupling coefficients.

5. CALCULATION OF COUPLING COEFFICIENTS FOR SHG

In order to calculate the coefficients we need only substitute the explicit form of the modes from Eq. (1.4) into Eqs. (4.4) and carry out the integration. $C_{mnrskl}^{2\omega}$ is then given by

$$C_{mnrskl}^{2\omega} = - \frac{8\sqrt{2}i\omega^2 d_{31} e^{i\Delta k z} (1+i\xi_2)^{(m+n)/2}}{k_2 c^2 (2^{m+n+r+s+k+l} m! n! r! s! k! l! \pi)^{1/2} (1-i\xi_2)^{(m+n)/2+1}} \times \frac{(1+i\xi_1)^{(r+s+k+l)/2} I_{mrk}^y I_{nsl}^x}{(1-i\xi_1)^{(r+s+k+l)/2+2} (\epsilon_z/\epsilon_x)^{1/4} w_{02} w_{01}^2}, \quad (5.1)$$

where the integrals I_{mrk}^y and I_{nsl}^x are given by

$$I_{mrk}^y = \int_{-\infty}^{\infty} dy H_m\left(\frac{y\sqrt{2}}{w_2}\right) H_r\left(\frac{y\sqrt{2}}{w_1}\right) H_k\left(\frac{y\sqrt{2}}{w_1}\right) \times \exp\left(-\frac{y^2}{w_{02}^2(1+i\xi_2)} - \frac{2y^2}{w_{02}^2(1-i\xi_1)}\right), \quad (5.2a)$$

$$I_{nsl}^x = \int_{-\infty}^{\infty} dx H_n\left(\left(\frac{\epsilon_z}{\epsilon_x}\right)^{1/2} \frac{x\sqrt{2}}{w_2}\right) H_s\left(\frac{x\sqrt{2}}{w_1}\right) H_l\left(\frac{x\sqrt{2}}{w_1}\right) \times \exp\left(-\frac{\epsilon_z x^2}{\epsilon_x w_{02}^2(1+i\xi_2)} - \frac{2x^2}{w_{01}^2(1-i\xi_1)}\right). \quad (5.2b)$$

In these equations the subscript 2 refers to quantities associated with the second-harmonic field, the subscript 1 to those of the fundamental field. The integrals (5.2) are essentially the same; they may be evaluated with the help of the generating function for Hermite polynomials

$$\exp(2sx - s^2) = \sum_{n=1}^{\infty} \frac{H_n(x)s^n}{n!}. \quad (5.3)$$

I_{mrk} will be the coefficient of $t^m s^r p^k$ multiplied by $m! r! k!$ in the expansion of the following integral in powers of t , s , and p :

$$I = \frac{w_1}{\sqrt{2}} \int_{-\infty}^{\infty} \exp[-b^2 u^2 + 2u(\sqrt{2}\alpha t + s + p) - t^2 - s^2 - p^2] du, \quad (5.4)$$

where

$$u = x\sqrt{2}/w_1, \quad \alpha = w_1/w_2\sqrt{2},$$

and

$$b^2 = \alpha^2(1-i\xi_2) + (1+i\xi_1).$$

Carrying out the integration, we have

$$I = (w_1/b)(\frac{1}{2}\pi)^{1/2} \times \exp[(\alpha\sqrt{2} + s + p)^2/b^2 - t^2 - s^2 - p^2]. \quad (5.5)$$

Expanding the exponential as a power series and using the binomial theorem, Eq. (5.5) may be written

in the form

$$I = \frac{w_1}{b} (\frac{1}{2}\pi)^{1/2} \sum_{n=0}^{\infty} \sum_{j=0}^n \sum_{i=0}^{2j} \sum_{q=n-j}^{i=2j} \sum_{l=u}^{q=n-j} \sum_{v=0}^{l=u} \frac{(-1)^{n-j}}{n!} \times \binom{n}{j} \binom{2j}{i} \binom{n-j}{q} \binom{u}{l} \binom{q}{v} \times b^{-2j} (\alpha\sqrt{2})^{2j-r} p^{l+2v} s^{i-l-2q-2v/2n-i-2q}. \quad (5.6)$$

Now the required coefficient will be given by the sum of the terms in this expansion, for which

$$\begin{aligned} 2n-i-2q &= n, \\ i-l-2q-2v &= r, \\ l+2v &= k. \end{aligned} \quad (5.7)$$

Adding these three equations, we obtain

$$n = \frac{1}{2}(m+r+k). \quad (5.8)$$

Therefore $m+r+k$ must be an even integer, which just expresses the symmetry conditions: An odd plus an even fundamental mode couples to an odd second-harmonic mode, an odd plus an odd fundamental mode couples to an even second-harmonic mode, and an even plus an even fundamental mode couples to an even second-harmonic mode.

The coefficients can be evaluated although the final expression is rather complicated in general. However, the low-order coefficients are simple in form; for example,

$$\begin{aligned} I_{000}^y &= (w_1/b)(\frac{1}{2}\pi)^{1/2}, \\ I_{020}^y &= (w_1/b)(\frac{1}{2}\pi)^{1/2}(1/b-1), \\ I_{101}^y &= (w_1/b)(\frac{1}{2}\pi)^{1/2}/2b. \end{aligned} \quad (5.9)$$

The most important set for which we will give an explicit expression are those coefficients resulting from the coupling between the lowest-order Gaussian fundamental mode and the $2n$ th second-harmonic mode:

$$I_{002n}^y = (w_1/b)(\frac{1}{2}\pi)^{1/2}(2\alpha^2/b^2-1)^n (2n)!/n!. \quad (5.10)$$

Substituting for α , b , and w from Eq. (5.4), and defining

$$w = w_{01}/w_{02}\sqrt{2}, \quad (5.11)$$

Eq. (5.10) becomes

$$I_{002n}^y = w_{01}(\frac{1}{2}\pi)^{1/2} \frac{(2n)! (1-i\xi_1)(1+i\xi_2)^{n+1/2}}{n! (1-i\xi_2)^n} \times \frac{[w^2-1-i(w^2\xi_1-\xi_2)]^n}{[w^2+1-i(w^2\xi_1-\xi_2)]^{n+1/2}}. \quad (5.12)$$

To obtain the final expression we substitute this expression together with that for I_{002m}^x into Eq. (5.2). I_{002m}^x is obtained from Eq. (5.12) by just substituting

$\bar{e}w$ for w , where $\bar{e} = (\epsilon_x/\epsilon_z)^{1/2}$ [see Eqs. (5.1)]:

$$C_{\{0\}2n2m}^{2\omega} = -\frac{4\sqrt{2}\pi^{1/2}i\omega^2 d_{31} e^{i\Delta k z} [(2m)!(2n)!]}{k_2 c^2 m! 2^m n! 2^n} \times \frac{[w^2 - 1 - i(w^2 \xi_1 - \xi_2)]^n [\bar{e}^2 w^2 - 1 - i(\bar{e}^2 w^2 \xi_1 - \xi_2)]^m}{\bar{e}^{1/2} w_{02} (1 - i\xi_1) [w^2 + 1 - i(w^2 \xi_1 - \xi_2)]^{n+1/2} [\bar{e}^2 w^2 + 1 - i(\bar{e}^2 w^2 \xi_1 - \xi_2)]^{m+1/2}}. \quad (5.13)$$

The factors $f = [(2n)!]^{1/2}/n! 2^n$ have values

n	0	1	2	3	4	6	10
f	1	0.7071	0.6724	0.5590	0.5229	0.4749	0.4197

and so decrease as n increases, reducing the coupling between the lowest-order fundamental mode and the $(2n, 2m)$ th second-harmonic mode with increasing n, m .

6. MISALIGNMENT

In calculating the expression for the coefficients (5.13) we have made the assumptions that the two beams propagate on the same axis and have coincident foci. If the two beams are misaligned in any way, the coupling will be reduced. Misalignment may occur in one of three ways: (i) The foci of the two beams do not lie in the same xy plane. (ii) The beam axes may be parallel but separated. (iii) The axes may be inclined at an angle to each other.

(i) The modification of the theory to include the first possibility is trivial. We consider the fundamental beam to be focused as before at $z=0$ but the $2n$ th harmonic beam to be focused now at $z=f$. Therefore ξ_2 becomes

$$\xi_2 = 2(z-f)/k_2 w_{02}^2, \quad (6.1)$$

and this does not affect the calculation of the coupling coefficients.

(ii) If the axis of the fundamental beam is removed from the z axis by a distance v in the y direction, the equation describing the m th mode is

$$\mathcal{E}_{mn} = \frac{\sqrt{2}}{w_{01}(2^{m+n} n! m! \pi)^{1/2}} \frac{(1+i\xi)^{(m+n)/2}}{(1-i\xi)^{(m+n)/2+1}} \times H_n\left(\frac{x\sqrt{2}}{w_1}\right) H_m\left(\frac{(y-v)\sqrt{2}}{w_1}\right) \times \exp\left(-\frac{[x^2 + (y-v)^2]}{w_{01}^2(1-i\xi_1)}\right). \quad (6.2)$$

With this modification, Eq. (5.4) becomes

$$I = \frac{w_1}{\sqrt{2}} \int_{-\infty}^{\infty} \exp\{-u^2 b^2 + 2u[\alpha\sqrt{2} + s + p + \zeta(1+i\xi_1)] - l^2 - s^2 - p^2\} dy, \quad (6.3)$$

where $\zeta = v\sqrt{2}/w_1$.

Carrying out the integration, we have

$$I = \frac{w_1}{b} \left(\frac{1}{2}\pi\right)^{1/2} \times \exp\left[-\zeta^2(1+i\xi_1)\left(1 - \frac{(1+i\xi_1)}{b}\right) + \frac{(\alpha\sqrt{2} + s + p)^2}{b^2} - l^2 - s^2 - p^2 + \frac{(\alpha\sqrt{2} + s + p)\zeta}{b}\right]. \quad (6.4)$$

Thus the separation of the axes has introduced two extra factors. The first factor is

$$F_1 = \exp\{-\zeta^2(1+i\xi_1)[1 - (1+i\xi_1)/b]\}, \quad (6.5)$$

which modifies each coupling coefficient since it is independent of l , s , and p . The modulus of this expression is, on substituting for b ,

$$F_1 = \exp(-\theta\zeta^2), \quad (6.6)$$

where

$$\theta = \frac{w^2(1+\xi_1^2)(w^2+1)}{(w^2+1)^2 + (\xi_1 w^2 - \xi_2)^2} > 0.$$

Thus the coupling between any two modes has an over-all exponential decrease with increasing separation of the axes, although, of course, there may be local increase.

The second factor,

$$F_2 = \exp[2(\alpha\sqrt{2} + s + p)\zeta/b], \quad (6.7)$$

has as the argument of the exponential function an expression which depends on an odd power of l , s , and p and therefore it will break the symmetry condition expressed by Eq. (5.8). Consider as an example I_{001}^2 , which is identically zero under the symmetry condition (5.8). Now it is given by

$$I_{001}^2 = \frac{w_1}{b} \left(\frac{1}{2}\pi\right)^{1/2} \times \exp\left(\frac{-\zeta^2 w^2(1+\xi_1^2)}{[w^2 + 1 - i(w^2 \xi_1 - \xi_2)]}\right) \frac{2\zeta(\alpha)^{1/2}}{b}. \quad (6.8)$$

Substituting for ζ , we have

$$|C_{010000}^{2\omega}| \propto |v/w_{01}| \exp(-2\theta v^2/w_{01}^2). \quad (6.9)$$

Thus this coefficient is zero at $v=0$ as expected and

increases with increasing separation to a maximum at

$$v = \pm w_{01}/2(\theta)^{1/2}, \quad (6.10)$$

and thereafter decreases asymptotically to zero. Since θ depends on z , this does not give immediately the separation of the axes for maximum coupling in a given crystal, except for the near-field case when ξ_1 and $\xi_2 \ll 1$ throughout the crystal.

(iii) For the case when the axis of the fundamental beam is tilted at a small angle θ to the axis of the second-harmonic beam in the xz plane, the equation representing the m th fundamental mode may be written¹⁶

$$\begin{aligned} \mathcal{E}_{mn}^{\omega} = & \frac{1}{w_{01}(2^{m+n}n!m!\pi)^{1/2}} \frac{(1+i\xi)^{(m+n)/2}}{(1-i\xi)^{(m+n)/2+1}} \\ & \times H_n\left(\frac{x\sqrt{2}}{w_1}\right) H_m\left(\frac{y\sqrt{2}}{w_1}\right) \\ & \times \exp\left(-\frac{x^2+y^2}{w_{01}^2(1-i\xi)} - ik\theta y\right). \end{aligned} \quad (6.11)$$

Following the previous argument, Eq. (5.5) becomes

$$\begin{aligned} I = (w_1/b)(\frac{1}{2}\pi)^{1/2} \exp[(\alpha\sqrt{2}+s+p)^2/b^2 \\ - \sqrt{2}ik\theta w_1(\alpha\sqrt{2}+s+p) \\ - k^2\theta^2 w_1^2 - t^2 - s^2 - p^2]. \end{aligned} \quad (6.12)$$

Again the modification introduces two new factors, one of which modifies each coefficient and the second of which breaks the symmetry condition (5.8). I_{001}^v has a form similar to Eq. (6.8):

$$I_{001}^v = -(w_1/b)(\frac{1}{2}\pi)^{1/2} \exp(-\frac{1}{2}k^2 w_1^2 \theta^2) 2ik\theta w_1 \alpha. \quad (6.13)$$

The maximum coupling occurs at $\theta = \pm 1/kw_1$. θ depends on z and so again this condition will not give immediately the maximum coupling, except when ξ_1 and $\xi_2 \ll 1$ throughout the crystal.

7. PARAMETRIC AMPLIFICATION

There are now three optical fields at frequencies ω_1 , ω_2 , and ω_3 , where $\omega_3 = \omega_1 + \omega_2$. We assume that each field satisfies the conditions such that it may be expanded in a set of resonator modes.

Again taking the example of lithium niobate, the relevant second-order polarization is given by^{8,15}

$$\begin{aligned} P_y^{\omega_1} &= d_{15} E_y^{\omega_2*} E_x^{\omega_3}, \\ P_y^{\omega_2} &= d_{15} E_y^{\omega_1*} E_x^{\omega_3}, \\ P_x^{\omega_3} &= d_{31} E_y^{\omega_1} E_y^{\omega_2}. \end{aligned} \quad (7.1)$$

The pump field ω_3 propagates as an extraordinary field and the signal ω_1 and idler ω_2 propagate as ordinary fields. Any other combination may be treated similarly.

¹⁶ H. Kogelnik, in *Proceedings of the Symposium on Quasi-Optics, New York, 1964* (Polytechnic Press, Brooklyn, N. Y., 1964), p. 333.

Expanding the fields in terms of the resonator modes and proceeding exactly as for SHG, we obtain the coupled-mode equations

$$dA_{mn}^{\omega_1}/dz = C_{mnrskl}^{\omega_1} A_{rs}^{\omega_2*} B_{kl}^{\omega_3}, \quad (7.2a)$$

$$dA_{mn}^{\omega_2}/dz = C_{mnrskl}^{\omega_2} A_{rs}^{\omega_1*} B_{kl}^{\omega_3}, \quad (7.2b)$$

$$dB_{mn}^{\omega_1}/dz = C_{mnrskl}^{\omega_3} A_{rs}^{\omega_1} A_{kl}^{\omega_2}, \quad (7.2c)$$

where

$$\begin{aligned} C_{mnrskl}^{\omega_1} = & -\frac{2\pi i \omega_1^2 d_{15} e^{-i\Delta k z}}{k_1 c^2} \\ & \times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{E}_{mn}^{\omega_1*} \mathcal{E}_{rs}^{\omega_2*} \mathcal{E}_{kl}^{\omega_3} dx dy, \end{aligned}$$

and

$$\Delta k = k_3 - k_1 - k_2. \quad (7.3)$$

For a lossless medium we have by the same argument as used for SHG

$$\eta_{\omega_1} C_{mnrskl}^{\omega_1} + \eta_{\omega_2} C_{mnrskl}^{\omega_2} = -\eta_{\omega_3} C_{mnrskl}^{\omega_3*}. \quad (7.4)$$

Note that this result does reduce to that expressed by Eq. (4.5) because of the factor $\frac{1}{2}$ in Eq. (4.1b) due to the $\omega_1 - \omega_2$ symmetry.⁴ The coupling coefficients may be calculated exactly as in the previous case. Now they will be complicated by the presence of three sets of beam parameters.

In the small-conversion approximation, the case studied by KB, only the Eqs. (7.2a) and (7.2b) need be considered. If also the approximation is now made that in the interaction of a lowest-order (Gaussian) mode pump beam with a Gaussian mode signal beam only the lowest-order modes of each field need be considered, Eqs. (7.2) reduce to

$$dA^{\omega_1}/dz = C^{\omega_1} B A^{\omega_2*}, \quad (7.5a)$$

$$dA^{\omega_2}/dz = C^{\omega_2} B A^{\omega_1*}. \quad (7.5b)$$

These equations may then be solved without further approximation. A detailed analysis will be given in a further paper.

8. EFFECT OF ABSORPTION AND PRESENCE OF RESONATOR

Both the presence of absorption in the uniaxial medium which for any medium of interest must necessarily be small and the presence of the resonator produce perturbation in the wave number k of the beam considered.

In the presence of absorption, k may be written

$$k = k' + ik'', \quad (8.1)$$

where $k'' \ll k'$.

The resonant frequencies of the open optical resonator are given by the equation

$$k_{q,m,n} = [q\pi + (m+n+1) \tan^{-1}(\psi)]/d, \quad (8.2)$$

where ψ and d are functions of the resonator, and q is an integer, the longitudinal-mode number; d is the

length of the resonator, and

$$\psi = [d(r_1 + r_2 - d)/(r_1 - d)(r_2 - d)]^{1/2}, \quad (8.3)$$

where r_1 and r_2 are the radii of curvature of the two end mirrors. For any physical situation we may assume $m, n < 100$ and hence $|k_{qmn} - k_{q00}| \ll k_{q00}$, for optical frequencies. In both cases, writing $k + \delta k$ in place of k , Eq. (1.3) becomes

$$\frac{\partial^2 \mathcal{E}}{\partial x^2} + \frac{\partial^2 \mathcal{E}}{\partial y^2} - 2i(k + \delta k) \frac{\partial \mathcal{E}}{\partial z} = 0. \quad (8.4)$$

To the approximations that have been used to obtain this equation, the term involving δk may be neglected. Hence the mode shape, and thus the foregoing theory, is unaltered by these modifications. These variations will, of course, appear in the term e^{-ikz} in each case.

9. CONCLUSION

A method has been developed with which nonlinear optical problems involving the interaction of the

traveling-wave form of the modes of the open optical resonator can be studied directly. In general, the coupling coefficients between the various modes may be calculated analytically and so their variation with the various parameters may be studied directly. Explicit expressions have been given for some of the important coefficients for SHG. From these general results, effects arising from strength and position of focusing can be obtained. It has been shown¹⁷ that the results for SHG are consistent with those found by other authors.^{6,9} These results will be published in further papers.

ACKNOWLEDGMENTS

I am very grateful to Dr. E. H. Hutten for help and encouragement during all the stages of this work, and I am also grateful to Dr. R. C. Smith and Dr. M. J. Colles of Southampton University for stimulating discussions.

¹⁷ R. Asby, Ph.D. thesis, University of London, 1968 (unpublished).