account, and that is the question of the distortion of the ionic wave functions due to the lattices. This question is difficult to answer at present, because of the complications encountered in any attempt to obtain self-consistency in a calculation of this magnitude.[18] However, preliminary investigations made by solving for a chlorine and an alkali ion in the Madelung field of the NaCl crystal indicate that the chlorine ion would be compressed in comparison to the free case and that the sodium ion is larger than in the free case. In this event, the more compact chlorine wave functions sample less of the detailed structure of the ions comprising the lattice, and hence one would expect the bands to narrow. That is, if one included a more accurate lattice potential in a tight-binding formalism, and also used wave functions obtained by solving the ion in the Madelung field, one would find the bands narrowing again. To this extent, the bands obtained in the present paper, including the complete lattice to a spherical approximation, could be considered as a determination of the maximum width of the NaCl valence band.

The author feels it would be useful to determine the bands by a combined use of tight binding and orthogonalized-plane-wave formalism[1,2] as this latter formalism treats the lattice potential in an essentially exact manner. It is felt that a use of an augmented-plane-wave model would not be totally satisfactory in that such a model requires that the crystal potential be spherically symmetric about a given nucleus in the region about that nucleus and that it be constant in all other regions.[7,8] Thus such a model would treat in a highly approximate manner regions in which the shape of the potential is seen to be important. In addition the augmented-plane-wave model has the defect of neglecting nonspherical parts of the potential in the regions around the nuclei.

[18] D. G. Shankland, Bull. Am. Phys. Soc. 11, 387 (1966).

V. ACKNOWLEDGMENTS

The author wishes to express his deepest appreciation to Professor W. J. Van Sciver for his advice and encouragement in this calculation. The author thanks Professor W. Beall Fowler and Donald Beck for several helpful discussions and the staff of the Courant Institute's computing center for their hospitality while these calculations were being performed there. Financial support by the U. S. Atomic Energy Commission is gratefully acknowledged.

PHYSICAL REVIEW VOLUME 162, NUMBER 3 15 OCTOBER 1967

Coupled-Wave Solution of Harmonic Generation in an Optically Active Medium*

PAUL P. BEY AND HERBERT RABIN
United States Naval Research Laboratory, Washington, D. C.
(Received 18 April 1967)

The equations governing the propagation and growth of coupled fields in a rotatory dispersive nonlinear medium are derived, making use of the method of Armstrong, Bloembergen, Ducuing, and Pershan. The results obtained for the intensity of harmonic radiation and the conditions for phase matching are compared to those obtained previously for propagation along the optic axis of crystals of point symmetry 32, and are found to be identical in the limiting case of nonattenuation of the fundamental wave. The coupled solutions for buildup of second-harmonic radiation are examined in the phase-matched case, and it is shown that the growth of circularly polarized harmonic light results from the depletion of the fundamental of opposite sense of circular polarization.

INTRODUCTION

It has been shown that new phase-matching conditions exist for harmonic generation in a nonlinear medium which is rotatory dispersive.[1] The treatment followed that of Franken and Ward[2] whereby the far-field harmonic-radiation intensity was obtained by summing the radiative contributions of the dipolar distribution in the medium produced by the incoming laser beam. This calculation was carried out in the limiting case in which the fundamental wave propagating through the medium was unattenuated, the so-called small-signal case.

It is of interest to extend this analysis to the more general case in which coupling between the fundamental and harmonic waves is taken account of making use of the method of Armstrong, Bloembergen, Ducuing, and Pershan.[3] Besides showing the manner in which rotatory dispersion can be fitted into the coupled-wave formalism, it is instructive to examine the predictions of the

* A preliminary report of this paper was presented at the Washington, D. C., meeting of the American Physical Society, 24 April 1967 [Bull. Am. Phys. Soc., 12, 578 (1967)].
[1] H. Rabin and P. P. Bey, Phys. Rev. 156, 1010 (1967).
[2] P. A. Franken and J. F. Ward, Rev. Mod. Phys. 35, 23 (1963).
[3] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).

coupled-wave solutions in an optically active medium and to compare the results in the appropriate limit to those obtained earlier employing the alternative Franken-Ward analysis.

As in the earlier treatment the cases of second-harmonic generation (SHG) and third-harmonic generation (THG) for propagation along the optic axis of uniaxial crystals of class 32 have been selected for evaluation, and the third-harmonic analysis is also appropriate to isotropic media. Both cases are evaluated in the limit of nonattenuation of the fundamental wave, and a solution of the coupled-wave equations for SHG is also obtained.

I. THEORY

The coupled-wave solution of harmonic generation is obtained from the wave equation containing a nonlinear source term. In the present problem where the nonlinear medium is taken to be rotatory dispersive, it is accordingly necessary that the wave equation be generalized to include optical rotation. This generalization follows from the treatment given by Landau and Lifshitz[4] of the propagation of waves through a linear optically active medium. It is instructive to present this linear treatment in order to provide the mathematical framework for the nonlinear problem. Optical rotation in a linear medium is carried out below in Sec. II, and in Sec. III the nonlinear problem is considered for the special cases of SHG and THG referred to in the Introduction.

II. OPTICAL ROTATION IN A LINEAR MEDIUM AFTER LANDAU AND LIFSHITZ

For an optically active medium the dielectric permeability tensor ϵ_{ij} is taken as a linear combination of the permeability tensor without rotatory dispersion $\epsilon_{ij}{}^{(0)}$ and terms involving the first-order spatial derivatives,

$$\epsilon_{ij}=\epsilon_{ij}{}^{(0)}+\gamma_{ijl}\frac{\partial}{\partial x_l}. \tag{1}$$

This second-rank tensor connects the electric displacement and field components in the usual manner

$$D_i=\epsilon_{ij}E_j, \tag{2}$$

where the summation convention applies to the three-dimensional suffixes. γ_{ijl} is a third-rank tensor, obeying the symmetry condition $\gamma_{ijl}=-\gamma_{jil}$ and for a nonabsorbing rotatory dispersive medium it is real. In those cases in which the propagation of a plane polarized wave is described by a single refractive index $(\epsilon^{(0)})^{1/2}$, for example isotropic media, cubic crystals, or propagation along the optic axis of doubly refracting crystals, γ_{ijl} reduces to the form

$$\gamma_{ijl}=(cf/\omega)e_{ijl}, \tag{3}$$

where c is the speed of light in vacuum, ω is the angular frequency of the propagating wave, e_{ijl} is the unit permutation tensor of third rank, and f is a pseudoscalar measuring the magnitude of specific rotation at frequency ω. Equation (3) is appropriate for the specific cases to be treated subsequently in the nonlinear problem.

Substitution of Eqs. (1) and (3) into Eq. (2) gives the expression for the transverse components of electric displacement,

$$D_i=\left[\epsilon^{(0)}\delta_{ij}+\left(\frac{cf}{\omega}\right)e_{ijl}\frac{\partial}{\partial x_l}\right]E_j, \tag{4}$$

which in turn is substituted into Maxwell's equations for a lossless dielectric medium. Assuming a plane-wave solution of the form $E_j(\omega,z)e^{-i\omega t}$, the transverse-electric-field components $E_x(\omega,z)$ and $E_y(\omega,z)$ at frequency ω are then described by the time-independent wave equation

$$\left[(\nabla^2+k^2)\delta_{ij}+\left(\frac{\omega f}{c}\right)e_{ijl}\frac{\partial}{\partial x_l}\right]E_j(\omega,z)=0, \tag{5}$$

where the field components depend only on one spatial coordinate z, the direction of propagation. The wave vector k is defined by the relation

$$k^2=\epsilon^{(0)}(\omega/c)^2. \tag{6}$$

Solutions of Eq. (5) yield two circularly polarized modes with opposite senses of rotation, and with different wave vectors. Four field components satisfying Eq. (5) can be written in shorthand notation

$$E_x{}^{\pm}(\omega,z)=\pm iE_y{}^{\pm}(\omega,z)=E_0e^{ik^{\pm}z}, \tag{7}$$

in which a given equation is written with either the upper or the lower of the indicated signs (+ and −). The constant factor E_0 is chosen as the amplitude common to both circularly polarized modes, k^+ is the wave vector associated with the left-handed circularly polarized wave with components $E_x{}^+$ and $E_y{}^+$, and k^- is correspondingly the wave vector of the right-handed wave with components $E_x{}^-$ and $E_y{}^-$. The wave equation requires that the above solutions are subject to the subsidiary conditions,

$$k^+-k^-=\omega f/c \tag{8}$$

and

$$k=(k^+k^-)^{1/2}\simeq\tfrac{1}{2}(k^++k^-); \tag{9}$$

the latter approximation holding in the case where $k^2\gg(\omega f/2c)^2$.

It is easily verified that Eq. (7) describes a plane-polarized wave of amplitude E_0 by defining

$$E_j(\omega,z)=\tfrac{1}{2}[E_j{}^+(\omega,z)+E_j{}^-(\omega,z)] \tag{10}$$

[4] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts 1960), p. 337.

for $j=x$ and y. Substitution gives the expressions

$$E_x(\omega,z)=E_0e^{ikz}\cos\alpha z \quad (11)$$

and

$$E_y(\omega,z)=E_0e^{ikz}\sin\alpha z\,, \quad (12)$$

corresponding to a rotating plane-polarized wave with specific rotation along the z direction given by

$$\alpha=\tfrac{1}{2}(k^+-k^-)=\omega f/2c\,. \quad (13)$$

The approximation of Eq. (9) is seen to require that $k^2 \gg \alpha^2$ the validity of which will be assumed throughout the following discussion.

III. OPTICAL ROTATION IN A NONLINEAR MEDIUM

The above treatment of optical rotation in a linear medium can now be extended to a rotatory-dispersive nonlinear medium in which harmonic waves are generated. We follow the method of Armstrong, Bloembergen, Ducuing, and Pershan[3] in treating the inhomogeneous wave equation obtained by the inclusion of a nonlinear polarization term. In the present problem the wave equation is generalized to include optical rotation of the propagating waves.

We consider coupling of the harmonic and fundamental waves along the z axis, and the frequency components are designated as ω_i. The x and y components of the field for a plane wave are written

$$E_j^{\pm}(\omega_i,z)e^{-i\omega_i t}\,, \quad (14)$$

and the corresponding nonlinear polarizations are

$$P_j^{\mathrm{NL}\pm}(\omega_i,z)e^{-i\omega_i t}\,. \quad (15)$$

These expressions lead to the following generalization of Eq. (5) for a nonlinear medium

$$\frac{\partial^2 E_x^{\pm}(\omega_i,z)}{\partial z^2}+k_i^2E_x^{\pm}(\omega_i,z)+\frac{\omega_i f_i}{c}\frac{\partial E_y^{\pm}(\omega_i,z)}{\partial z} =4\pi\left(\frac{\omega_i}{c}\right)^2 P_x^{\mathrm{NL}\pm}(\omega_i,z)\,, \quad (16)$$

and

$$\frac{\partial^2 E_y^{\pm}(\omega_i,z)}{\partial z^2}+k_i^2E_y^{\pm}(\omega_i,z)-\frac{\omega_i f_i}{c}\frac{\partial E_x^{\pm}(\omega_i,z)}{\partial z} =4\pi\left(\frac{\omega_i}{c}\right)^2 P_y^{\mathrm{NL}\pm}(\omega_i,z)\,. \quad (17)$$

We have anticipated in Eq. (14) both right-handed and left-handed circularly polarized field variables (designated $-$ and $+$, respectively) in accord with the earlier solutions of Eq. (5). Furthermore, right-handed and left-handed modes for the nonlinear polarization, Eq. (15), are also assumed. Explicit forms of these polarizations will subsequently be derived for the specific cases to be considered. It should be noted that in Eqs. (16) and (17) a particular sense of rotation of the nonlinear polarization is taken as the generator of fields with the corresponding sense of rotation. The wave vector k_i associated with the frequency ω_i is defined in parallel form to Eq. (6)

$$k_i^2=\epsilon^{(0)}(\omega_i/c)^2\,. \quad (18)$$

Also in Eqs. (16) and (17) we admit the possibility that the quantity f is dispersive, and we associate f_i with a particular ω_i.

We now look for solutions of the inhomogeneous wave equations of the form

$$E_x^{\pm}(\omega_i,z)=\pm iE_y^{\pm}(\omega_i,z)=E_{\omega_i}^{\pm}(z)e^{ik_i^{\pm}z}\,, \quad (19)$$

where $E_{\omega_i}^{-}(z)$ and $E_{\omega_i}^{+}(z)$ are the complex amplitudes of the right-handed and left-handed circularly polarized waves at frequency ω_i. These amplitudes are a function of the coordinate z allowing for growth or decay of the propagating waves. In other respects the above expression is analogous to the former solution, Eq. (7) of the homogeneous wave equation, with the requirement from Eqs. (9) and (13) that

$$k_i=(k_i^+k_i^-)^{1/2}\simeq\tfrac{1}{2}(k_i^++k_i^-) \quad (20)$$

and

$$\alpha_i=\tfrac{1}{2}(k_i^+-k_i^-)=\omega_i f_i/2c\,. \quad (21)$$

We also define

$$E_j(\omega_i,z)=\tfrac{1}{2}[E_j^+(\omega_i,z)+E_j^-(\omega_i,z)] \quad (22)$$

in an analogous manner to Eq. (10), and the components of nonlinear polarization at frequency ω_i are written in terms of right-handed and left-handed circular forms,

$$P_x^{\mathrm{NL}\pm}(\omega_i,z)=\pm iP_y^{\mathrm{NL}\pm}(\omega_i,z)\,. \quad (23)$$

Upon applying Eqs. (19)–(21) and (23) to the wave equations (16) and (17), the simplified form is obtained

$$\frac{dE_{\omega_i}^{\pm}(z)}{dz}=\frac{2\pi i}{k_i}\left(\frac{\omega_i}{c}\right)^2 P_x^{\mathrm{NL}\pm}(\omega_i,z)e^{-ik_i^{\pm}z}\,, \quad (24)$$

where it is assumed that the term involving the second-order derivative is small compared to the term containing the first-order derivative and can be ignored.

We now examine this equation for specific cases of SHG and THG. Phase-matching conditions are derived and the nature of the solution is examined in the small-signal limit, and in the case of SHG when coupling between the fundamental and harmonic waves is taken into account.

A. SHG along the Optic Axis of Crystals of Point Symmetry 32

The axis of a crystal of symmetry class 32 is chosen so that the z axis corresponds to the threefold or optic axis and the x axis is a twofold axis. The wave vector of the incident wave is assumed to be directed along the positive z axis, and it is further assumed that the incident wave is linearly polarized along the x axis upon entering the crystal face oriented along the $z=0$ plane. The latter can be assured from Eqs. (19) and (22) by requiring

$$E_{\omega_1}{}^{+}(0)=E_{\omega_1}{}^{-}(0), \tag{25}$$

where ω_1 is taken as the frequency of the fundamental wave. Writing ω_2 for the second-harmonic frequency

$$\omega_2=2\omega_1, \tag{26}$$

it is assumed that the $z=0$ boundary condition on the second-harmonic wave is

$$E_{\omega_2}{}^{+}(0)=E_{\omega_2}{}^{-}(0)=0. \tag{27}$$

We now consider the resultant field in the medium produced by the superposition of the fundamental and harmonic waves, given by the expression

$$E_j{}^{\pm}(\omega_1,\omega_2,z)=E_j{}^{\pm}(\omega_1,z)+E_j{}^{\pm}(\omega_2,z), \tag{28}$$

where as before j refers to the x and y coordinates. Defining the real quantities

$$\mathcal{E}_j{}^{\pm}=\mathrm{Re}E_j{}^{\pm}(\omega_1,\omega_2,z) \tag{29}$$

and substituting from Eq. (19), we have

$$\begin{aligned}E_x{}^{\pm}(\omega_1,\omega_2,z)&=\pm iE_y{}^{\pm}(\omega_1,\omega_2,z)\\&=E_{\omega_1}{}^{\pm}(z)e^{ik_1{}^{\pm}z}+E_{\omega_2}{}^{\pm}(z)e^{ik_2{}^{\pm}z}.\end{aligned} \tag{30}$$

The associated second-order nonlinear polarization components written as real quantities, are

$$\mathcal{P}_x{}^{\mathrm{NL}}=d_{11}(\mathcal{E}_x{}^2-\mathcal{E}_y{}^2) \tag{31}$$

and

$$\mathcal{P}_y{}^{\mathrm{NL}}=-2d_{11}\mathcal{E}_x\mathcal{E}_y, \tag{32}$$

where d_{11} is the appropriate second-order susceptibility tensor for crystal class 32 for propagation along the optic axis,[2] and where

$$\mathcal{E}_j=\tfrac{1}{2}(\mathcal{E}_j{}^{+}+\mathcal{E}_j{}^{-}). \tag{33}$$

It is next desired to obtain the expression for the complex nonlinear polarization amplitude $P_x{}^{\mathrm{NL}\pm}(\omega_i,z)$ appearing in Eq. (24). The procedure is as follows. We write

$$\mathcal{E}_j=\mathrm{Re}E_j(\omega_1,\omega_2,z)=\tfrac{1}{2}[E_j(\omega_1,\omega_2,z)+(E_j(\omega_1,\omega_2,z))^*], \tag{34}$$

where

$$E_j(\omega_1,\omega_2,z)=\tfrac{1}{2}[E_j{}^{+}(\omega_1,\omega_2,z)+E_j{}^{-}(\omega_1,\omega_2,z)]. \tag{35}$$

The quantities $\mathcal{P}_j{}^{\mathrm{NL}}$ in Eqs. (31) and (32) are evaluated on the basis of the definitions given in Eqs. (30), (34) and (35). These polarizations show terms which are associated with dc effects and frequency-dependent effects in ω_1, ω_2 and higher harmonics. We are only interested in the present problem in terms associated with ω_1 and ω_2, and we designate $\mathcal{P}_j{}^{\mathrm{NL}}(\omega_1,z)$ to be that portion of $\mathcal{P}_j{}^{\mathrm{NL}}$ which is associated with ω_1, and similarly $\mathcal{P}_j{}^{\mathrm{NL}}(\omega_2,z)$ is that portion associated with ω_2. We thus have the expressions

$$\begin{aligned}\mathcal{P}_x{}^{\mathrm{NL}}(\omega_1,z)=\tfrac{1}{4}d_{11}\{[(E_{\omega_1}{}^{+}(z))^*E_{\omega_2}{}^{-}(z)e^{i(k_2{}^{-}-k_1{}^{+})z}\\+E_{\omega_2}{}^{+}(z)(E_{\omega_1}{}^{-}(z))^*e^{i(k_2{}^{+}-k_1{}^{-})z}]+\text{c.c.}\},\end{aligned} \tag{36}$$

$$\begin{aligned}\mathcal{P}_x{}^{\mathrm{NL}}(\omega_2,z)=\tfrac{1}{8}d_{11}\{[(E_{\omega_1}{}^{+}(z))^2e^{2ik_1{}^{+}z}\\+(E_{\omega_1}{}^{-}(z))^2e^{2ik_1{}^{-}z}]+\text{c.c.}\},\end{aligned} \tag{37}$$

$$\begin{aligned}\mathcal{P}_y{}^{\mathrm{NL}}(\omega_1,z)=\tfrac{1}{4}id_{11}\{[(E_{\omega_1}{}^{-}(z))^*E_{\omega_2}{}^{+}(z)e^{i(k_2{}^{+}-k_1{}^{-})z}\\-(E_{\omega_1}{}^{+}(z))^*E_{\omega_2}{}^{-}(z)e^{i(k_2{}^{-}-k_1{}^{+})z}]+\text{c.c.}\},\end{aligned} \tag{38}$$

and

$$\begin{aligned}\mathcal{P}_y{}^{\mathrm{NL}}(\omega_2,z)=\tfrac{1}{8}id_{11}\{[(E_{\omega_1}{}^{+}(z))^2e^{2ik_1{}^{+}z}\\-(E_{\omega_1}{}^{-}(z))^2e^{2ik_1{}^{-}z}]+\text{c.c.}\}.\end{aligned} \tag{39}$$

It is now possible to identify the complex functions $P_j{}^{\mathrm{NL}}(\omega_1,z)$ and $P_j{}^{\mathrm{NL}}(\omega_2,z)$ by comparing the expressions

$$\mathcal{P}_j{}^{\mathrm{NL}}(\omega_1,z)=\tfrac{1}{2}[P_j{}^{\mathrm{NL}}(\omega_1,z)+(P_j{}^{\mathrm{NL}}(\omega_1,z))^*] \tag{40}$$

and

$$\mathcal{P}_j{}^{\mathrm{NL}}(\omega_2,z)=\tfrac{1}{2}[P_j{}^{\mathrm{NL}}(\omega_2,z)+(P_j{}^{\mathrm{NL}}(\omega_2,z))^*] \tag{41}$$

with Eqs. (36)–(39). We have then

$$\begin{aligned}P_x{}^{\mathrm{NL}}(\omega_1,z)=\tfrac{1}{2}d_{11}[(E_{\omega_1}{}^{+}(z))^*E_{\omega_2}{}^{-}(z)e^{i(k_2{}^{-}-k_1{}^{+})z}\\+E_{\omega_2}{}^{+}(z)(E_{\omega_1}{}^{-}(z))^*e^{i(k_2{}^{+}-k_1{}^{-})z}],\end{aligned} \tag{42}$$

$$\begin{aligned}P_x{}^{\mathrm{NL}}(\omega_2,z)=\tfrac{1}{4}d_{11}[(E_{\omega_1}{}^{+}(z))^2e^{2ik_1{}^{+}z}\\+(E_{\omega_1}{}^{-}(z))^2e^{2ik_1{}^{-}z}],\end{aligned} \tag{43}$$

$$\begin{aligned}P_y{}^{\mathrm{NL}}(\omega_1,z)=\tfrac{1}{2}id_{11}[(E_{\omega_1}{}^{-}(z))^*E_{\omega_2}{}^{+}(z)e^{i(k_2{}^{+}-k_1{}^{-})z}\\-(E_{\omega_1}{}^{+}(z))^*E_{\omega_2}{}^{-}(z)e^{i(k_2{}^{-}-k_1{}^{+})z}],\end{aligned} \tag{44}$$

and

$$\begin{aligned}P_y{}^{\mathrm{NL}}(\omega_2,z)=\tfrac{1}{4}id_{11}[(E_{\omega_1}{}^{+}(z))^2e^{2ik_1{}^{+}z}\\-(E_{\omega_1}{}^{-}(z))^2e^{2ik_1{}^{-}z}].\end{aligned} \tag{45}$$

By inspection these equations yield the circularly polarized forms of nonlinear polarization

$$\begin{aligned}P_x{}^{\mathrm{NL}\pm}(\omega_1,z)&=\pm iP_y{}^{\mathrm{NL}\pm}(\omega_1,z)\\&=\tfrac{1}{2}d_{11}(E_{\omega_1}{}^{\pm}(z))^*E_{\omega_2}{}^{\mp}(z)e^{i(k_2{}^{\mp}-k_1{}^{\pm})z}\end{aligned} \tag{46}$$

and

$$\begin{aligned}P_x{}^{\mathrm{NL}\pm}(\omega_2,z)&=\pm iP_y{}^{\mathrm{NL}\pm}(\omega_2,z)\\&=\tfrac{1}{4}d_{11}(E_{\omega_1}{}^{\mp}(z))^2e^{2ik_1{}^{\mp}z},\end{aligned} \tag{47}$$

which are recognized as the assumed forms given in Eq. (23).

Upon substituting Eqs. (46) and (47) into Eq. (24) the following two sets of coupled-amplitude equations are obtained:

$$\frac{dE_{\omega_1}{}^{\pm}(z)}{dz}=\frac{i\pi d_{11}}{k_1}\left(\frac{\omega}{c}\right)^2(E_{\omega_1}{}^{\pm}(z))^*\times E_{\omega_2}{}^{\mp}(z)e^{i[(\Delta k)_{\mathrm{SHG}}\mp(\Delta\alpha)_{\mathrm{SHG}}]z} \tag{48}$$

and

$$\frac{dE_{\omega_2}{}^{\pm}(z)}{dz}=\frac{i2\pi d_{11}}{k_2}\left(\frac{\omega}{c}\right)^2\times(E_{\omega_1}{}^{\mp}(z))^2e^{-i[(\Delta k)_{\mathrm{SHG}}\pm(\Delta\alpha)_{\mathrm{SHG}}]z}, \tag{49}$$

where we define

$$(\Delta k)_{\rm SHG}=k_2-2k_1, \tag{50}$$

$$(\Delta\alpha)_{\rm SHG}=\alpha_2+2\alpha_1, \tag{51}$$

and from Eqs. (20) and (21) we have used the identity $k_i^{\pm}=k_i\pm\alpha_i$ and, from Eq. (26), $\omega_2=2\omega_1=2\omega$.

It is now a simple matter to examine the harmonic amplitudes in the small-signal case where the fundamental amplitude is unattenuated in traversing the medium which we take to extend from $z=0$ to $z=l$. Equation (49) is directly integrated to yield the left-handed and right-handed second-harmonic amplitudes at the exit face of the medium,

$$E_{\omega_2}{}^{+}(l)=\frac{2\pi d_{11}}{k_2}\left(\frac{\omega}{c}\right)^2(E_{\omega_1}{}^{-}(0))^2\times\left\{\frac{1-e^{-i[(\Delta k)_{\rm SHG}+(\Delta\alpha)_{\rm SHG}]l}}{(\Delta k)_{\rm SHG}+(\Delta\alpha)_{\rm SHG}}\right\} \tag{52}$$

and

$$E_{\omega_2}{}^{-}(l)=\frac{2\pi d_{11}}{k_2}\left(\frac{\omega}{c}\right)^2(E_{\omega_1}{}^{+}(0))^2\times\left\{\frac{1-e^{-i[(\Delta k)_{\rm SHG}-(\Delta\alpha)_{\rm SHG}]l}}{(\Delta k)_{\rm SHG}-(\Delta\alpha)_{\rm SHG}}\right\}, \tag{53}$$

where the boundary condition stated in Eq. (27) is employed, and we have written $E_{\omega_1}{}^{\pm}(z)=E_{\omega_1}{}^{\pm}(0)$, a constant independent of z. The intensity of second-harmonic radiation at the exit face of the nonlinear medium is given by the following proportionality

$$I_{\rm SHG}\propto E_{\omega_2}{}^{+}(l)(E_{\omega_2}{}^{+}(l))^*+E_{\omega_2}{}^{-}(l)(E_{\omega_2}{}^{-}(l))^*. \tag{54}$$

Substituting the amplitudes from Eqs. (52) and (53) and employing Eq. (25) it follows that

$$I_{\rm SHG}\propto\frac{\sin^2\{\frac{1}{2}l[(\Delta k)_{\rm SHG}-(\Delta\alpha)_{\rm SHG}]\}}{[(\Delta k)_{\rm SHG}-(\Delta\alpha)_{\rm SHG}]^2}+\frac{\sin^2\{\frac{1}{2}l[(\Delta k)_{\rm SHG}+(\Delta\alpha)_{\rm SHG}]\}}{[(\Delta k)_{\rm SHG}+(\Delta\alpha)_{\rm SHG}]^2}. \tag{55}$$

Two phase-matching conditions

$$(\Delta k)_{\rm SHG}=\pm(\Delta\alpha)_{\rm SHG} \tag{56}$$

are apparent from Eq. (55).

In the case of perfect phase matching, the coupled-wave equations also have a solution which describes the depletion of the fundamental wave at the expense of the growth of the harmonic. Here we obtain a solution in a manner analogous to that given by Bloembergen[5] by choosing the amplitudes of ω_1 to be completely real quantities:

$$E_{\omega_1}{}^{\pm}(z)=\rho_{\omega_1}{}^{\pm}(z), \tag{57}$$

and the amplitudes of ω_2 to be completely imaginary:

$$E_{\omega_2}{}^{\pm}(z)=i\rho_{\omega_2}{}^{\pm}(z). \tag{58}$$

First, the phase-matching condition $(\Delta k)_{\rm SHG}=-(\Delta\alpha)_{\rm SHG}$ is considered. Upon substituting Eqs. (57) and (58) into Eqs. (48) and (49), the following coupled equations are obtained:

$$\frac{d\rho_{\omega_1}{}^{-}(z)}{dz}=-\frac{K}{k_1}\rho_{\omega_1}{}^{-}(z)\rho_{\omega_2}{}^{+}(z) \tag{59}$$

and

$$\frac{d\rho_{\omega_2}{}^{+}(z)}{dz}=\frac{2K}{k_2}(\rho_{\omega_1}{}^{-}(z))^2, \tag{60}$$

where $K=\pi d_{11}(\omega/c)^2$. Multiplying Eq. (59) by $(2/k_2)\rho_{\omega_1}{}^{-}(z)$ and Eq. (60) by $(1/k_1)\rho_{\omega_2}{}^{+}(z)$, and adding, it follows after integration that

$$(\rho_{\omega_1}{}^{-}(z))^2=(\rho_{\omega_1}{}^{-}(0))^2-\frac{k_2}{2k_1}(\rho_{\omega_2}{}^{+}(z))^2, \tag{61}$$

where the boundary conditions stated in Eqs. (25) and (27) have been used. [It can be shown that Eq. (61) deviates from the expression for the power-flow integral

$$(\rho_{\omega_1}{}^{-}(z))^2+(\rho_{\omega_2}{}^{+}(z))^2=(\rho_{\omega_1}{}^{-}(0))^2$$

by terms of the order of the approximation inherent in Eq. (24), which in turn are traceable to the approximate equality in Eq. (20).] Substituting Eq. (61) into Eq. (60) and integrating we have

$$\rho_{\omega_2}{}^{+}(z)=\left(\frac{2k_1}{k_2}\right)^{\frac{1}{2}}\rho_{\omega_1}{}^{-}(0)\tanh\left[K\left(\frac{2}{k_1k_2}\right)^{\frac{1}{2}}\rho_{\omega_1}{}^{-}(0)z\right] \tag{62}$$

and

$$\rho_{\omega_1}{}^{-}(z)=\rho_{\omega_1}{}^{-}(0)\,\mathrm{sech}\left[K\left(\frac{2}{k_1k_2}\right)^{1/2}\rho_{\omega_1}{}^{-}(0)z\right]. \tag{63}$$

By a completely analogous treatment for the other phase-matched condition, $(\Delta k)_{\rm SHG}=(\Delta\alpha)_{\rm SHG}$, it can similarly be shown that

$$\rho_{\omega_2}{}^{-}(z)=\left(\frac{2k_1}{k_2}\right)^{1/2}\rho_{\omega_1}{}^{+}(0)\tanh\left[K\left(\frac{2}{k_1k_2}\right)^{1/2}\rho_{\omega_1}{}^{+}(0)z\right] \tag{64}$$

and

$$\rho_{\omega_1}{}^{+}(z)=\rho_{\omega_1}{}^{+}(0)\,\mathrm{sech}\left[K\left(\frac{2}{k_1k_2}\right)^{1/2}\rho_{\omega_1}{}^{+}(0)z\right]. \tag{65}$$

The significance of these results will be left for the discussion section. We now turn our attention to the case of THG.

[5] N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965), pp. 88–89.

B. THG along the Optic Axis of Crystals of Point Symmetry 32 and THG in Isotropic Media

We next calculate the intensity of third-harmonic radiation generated by a wave propagating along the optic axis of a crystal of class 32. The orientation of coordinate axes is the same as that defined above in Sec. IIIA, and the x and y components of third-order nonlinear polarization are given by

$$\mathcal{P}_j^{\rm NL}=3C_{1122}\mathcal{E}_j(\mathcal{E}_x^2+\mathcal{E}_y^2)\,, \qquad (66)$$

where C_{1122} is the nonvanishing component of the fourth-rank electric-susceptibility tensor.[6] Equation (66) is an expression involving real field and polarization quantities and in the notation adopted these quantities are written in script symbols. As was pointed out earlier[1] the form of Eq. (66) is equally applicable for an isotropic medium.

The procedure is now identical to that carried out above in Sec. IIIA. Equation (25), (27)–(30), and (33)–(35) hold as before, with the exception that ω_2 is now replaced by the third-harmonic frequency, ω_3, and similarly k_3 replaces k_2. Equation (26) becomes $\omega_3=3\omega_1$. We repeat the procedure leading to the complex nonlinear polarizations. Upon substitution into Eq. (24) the following coupled-amplitude equations are obtained:

$$\frac{dE_{\omega_3}^{\pm}(z)}{dz}=\frac{i27\pi c_{1122}}{4k_3}\left(\frac{\omega^2}{c}\right)\Big[(E_{\omega_1}^{\pm}(z))^2E_{\omega_1}^{\mp}(z)\times e^{-i[(\Delta k)_{\rm THG}\pm(\Delta\alpha)_{\rm THG}]z}+\text{terms of the order of } E_{\omega_1}^2E_{\omega_3} \text{ and } E_{\omega_3}^3\Big], \qquad (67)$$

where we define

$$(\Delta k)_{\rm THG}=k_3-3k_1\,, \qquad (68)$$

$$(\Delta\alpha)_{\rm THG}=\alpha_3-\alpha_1\,, \qquad (69)$$

and

$$\omega_3=3\omega_1=3\omega\,.$$

We can now examine the third-harmonic amplitude in the small-signal case. Equations (67) are directly integrated by dropping the indicated terms of order $E_{\omega_1}^2E_{\omega_3}$ and $E_{\omega_3}^3$ in comparison to the leading $E_{\omega_1}^3$ term. Assuming the medium extends from $0\leq z\leq l$, the left-handed and right-handed third-harmonic amplitudes at the exit face are given by

$$E_{\omega_3}^{\pm}(l)=\frac{27\pi c_{1122}}{4k_3}\left(\frac{\omega}{c}\right)^2(E_{\omega_1}^{\pm}(0))^2E_{\omega_1}^{\mp}(0)\times\left\{\frac{1-e^{-i[(\Delta k)_{\rm THG}\pm(\Delta\alpha)_{\rm THG}]l}}{(\Delta k)_{\rm THG}\pm(\Delta\alpha)_{\rm THG}}\right\}, \qquad (70)$$

where we have written $E_{\omega_1}^{\pm}(z)=E_{\omega_1}^{\pm}(0)$. Assuming $E_{\omega_1}^{+}(0)=E_{\omega_1}^{-}(0)$, the intensity of third-harmonic radiation is given by

$$I_{\rm THG}\propto\frac{\sin^2\{\frac{1}{2}l[(\Delta k)_{\rm THG}-(\Delta\alpha)_{\rm THG}]\}}{[(\Delta k)_{\rm THG}-(\Delta\alpha)_{\rm THG}]^2}+\frac{\sin^2\{\frac{1}{2}l[(\Delta k)_{\rm THG}+(\Delta\alpha)_{\rm THG}]\}}{[(\Delta k)_{\rm THG}+(\Delta\alpha)_{\rm THG}]^2}, \qquad (71)$$

upon substituting Eq. (70) into the right hand side of Eq. (54) in which ω_2 is replaced by ω_3. Again two phase-matching conditions are apparent from Eq. (71),

$$(\Delta k)_{\rm THG}=\pm(\Delta\alpha)_{\rm THG}\,. \qquad (72)$$

IV. DISCUSSION

This calculation has shown that when a linearly polarized fundamental wave propagates through a nonlinear medium which is rotatory dispersive, two phase-matching conditions occur in the harmonic-generation process. These conditions are in contrast to the single phase-matching condition which typically occurs in the absence of rotation of the plane of polarization. In principle, the rotation of the plane of polarization may occur either by natural optical activity of the medium or by Faraday rotation, and the rotatory dispersion may be normal or anomalous. In the present treatment it was assumed that the medium was nonabsorbing at the frequencies of interest.

A primary objective of this paper was to compare results obtained earlier[1] based on the method of Franken and Ward with the coupled-wave method developed by Armstrong, Bloembergen, Ducuing, and Pershan. The Franken-Ward treatment is carried out in the limiting case in which the fundamental wave is assumed to be essentially nonattenuated; the fundamental sets up a dipolar array at the harmonic frequency and the problem reduces to computing the resultant harmonic radiation emanating from this array. The intensity functions and phase-matching conditions derived on this basis are given in Eqs. (23) and (24) for SHG and Eqs. (40) and (41) for THG in Ref. 1. These expressions are identical to corresponding results obtained in the present paper in Eqs. (55) and (56) and (71) and (72), where the coupled-wave equations were evaluated in the limit of nonattenuation of the fundamental. This agreement not only serves to confirm the earlier analysis, but tends to put both treatments on an equal footing for the analysis of problems in the limiting small-signal case. These alternative approaches each offer certain advantages in the elucidation of the harmonic generation problem.

In the present paper, Sec. IIIA, we have given a particular solution of the coupled-wave equations for phase-matched SHG along the optic axis of crystals of class 32. This solution describes the buildup of the harmonic wave at the expense of the fundamental. It is of particular interest that the right-handed and left-

[6] P. D. Maker and R. W. Terhune, Phys. Rev. 137, A801 (1965).

handed circularly polarized components of the fundamental wave act independently of one another. Each fundamental component couples to the harmonic component of opposite sense of circular polarization to yield coupled-amplitude equations analogous to the nonrotatory case.[3,5] Equations (62) and (63) show, for example in the phase-matched case $(\Delta k)_{\text{SHG}} = -(\Delta\alpha)_{\text{SHG}}$, the left-handed second-harmonic amplitude $\rho_{\omega_2}{}^{+}(z)$ grows as the hyperbolic tangent of z as the right-handed fundamental amplitude $\rho_{\omega_1}{}^{-}(z)$ decreases as the hyperbolic secant of z. Equations (64) and (65) indicate the corresponding result for opposite senses of circular polarization for the $(\Delta k)_{\text{SHG}} = (\Delta\alpha)_{\text{SHG}}$ phase-matching case. Thus, complete conversion to second harmonic could in principle only be attained in an optically active medium when the incoming fundamental wave is completely circularly polarized. It should be mentioned that the above remarks pertaining to coupling between opposite components of circular polarization in the coupled-wave solution also apply in the small-signal case as given in Eqs. (52) and (53). We see for example, that a circular polarized fundamental wave yields a second harmonic of opposite circular polarization.

The third-harmonic problem differs in several significant respects from that of second harmonic. The coupled-wave equations for THG, Eqs. (67), contain additional terms associated with the quadratic Kerr effect, and these are described as reactive terms which alter the phase velocities of the propagating waves.[3] It is of interest to mention that the effect of optical rotation is to modify certain of the Kerr terms such that they are transposed from purely reactive terms to terms which directly alter the power flow of third-harmonic radiation. The detailed nature of these terms will be presented in a later communication.

There is also a unique difference between the second- and third-harmonic cases in relation to the polarization of the fundamental wave. We have seen that in SHG the circularly polarized components act independently of one another in producing second-harmonic radiation. This simple separation of circularly polarized components apparently does not occur in the more complex coupled equations for THG. In the small-signal solution for THG given in Eqs. (70) both senses of circular polarization of the fundamental wave must be nonvanishing to yield a nonzero third-harmonic amplitude. This is basically traceable to the $(\mathcal{E}_x{}^2 + \mathcal{E}_y{}^2)$ factor in the expression for third-order nonlinear polarization, Eq. (66). It is clear that this factor is constant for pure circularly polarized fundamental radiation, and hence, a circularly polarized fundamental wave is unable to generate a 3ω nonlinear source term. This is not the case for linearly polarized fundamental radiation as we have shown above and in earlier work. These conclusions could easily be tested in a third-harmonic experiment, for example, in an isotropic medium or in quartz (along the optic axis) by suitable transformation of the fundamental wave from linear to circular polarization. There is no third-harmonic signal generated through the third-order nonlinear polarization with circularly polarized fundamental radiation, and the fundamental wave is modified only by quadratic Kerr terms. It is thus possible with circularly polarized light to produce pure quadratic Kerr effect without simultaneous third-harmonic generation.

In conclusion, there are several extensions of the present work which are indicated.

First, the present analysis can be extended to other optically active nonlinear media. The general features of the results would be expected to be largely unchanged. For example, in SHG different phase-matching conditions would be expected for the two senses of circular polarization with correspondingly different coherence lengths for each. The specific form of the nonlinear polarization term can lead to interesting results, as has been noted with respect to Eq. (66). There are undoubtedly other symmetry classes where the form of the nonlinear polarization is such as to produce unique effects depending on the polarization of the propagating waves.

Second, in addition to harmonic generation other nonlinear processes in rotatory dispersive media might be profitably investigated, such as frequency mixing, parametric amplification, self-focusing, etc.

Third, as has been indicated above the analysis for magnetooptic rotation such as in the Faraday effect is in principle analogous to that of natural optical activity. Faraday rotation, because of the obvious possibilities for tuning in nonlinear processes, would appear to offer a particularly fertile area of theoretical and experimental investigation.

Note added in proof. N. Bloembergen and J. Simon [Bull. Am. Phys. Soc. **12**, 687 (1967)] have reported on experimental studies of second harmonic generation in optically active crystals of symmetry class 23. For propagation along the threefold axis the form of the second order polarization is identical to that given in Eqs. (31)–(32) for symmetry class 32. Bloembergen and Simon have confirmed that the sense of circular polarization of the fundamental is opposite to that of the second harmonic, and there are different coherence lengths for the two senses of circular polarization. In a private communication, Professor Bloembergen has indicated similar unpublished findings for quartz with symmetry 32.

ACKNOWLEDGMENTS

The authors are indebted to Dr. F. T. Byrne and Dr. W. J. Condell of the Office of Naval Research for a helpful discussion during the preparation of this paper.