

ON THE THEORY OF NONLINEAR OPTICS AND DEVICES

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ABSTRACT

This thesis is concerned with some aspects of the theory of parametric interactions which have not been fully considered in the literature. The existing theory of nonlinear interactions in nonabsorbing, non-gyrotropic, anisotropic crystals has been generalised so that the basic theory of interactions in absorbing and gyrotropic crystals is now understood. Full allowance has been made for the fact that the interacting waves are elliptically polarized and not linearly polarized as they are in the nonabsorbing case. When an experiment has been performed on an absorbing or gyrotropic crystal to measure the nonlinear coefficients this theory must be used to obtain the nonlinear coefficient from the experimental results.

The equations governing the amplitudes of the interacting waves is found to be of the same form as for nonabsorbing non-gyrotropic crystals.

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I INTRODUCTION

The work which is described in this thesis has been performed in connection with the laser research group, which is a purely experimental group.

The theoretical subjects considered were chosen to augment the group's activities and to overcome limitations in the theory of nonlinear interactions and devices. The experimental work in the laser group is on three topics;

1. parametric amplification and oscillation in tellurium
2. parametric amplification and oscillation in proustite
3. second harmonic generation in tellurium with a view to measuring accurately the nonlinear coefficient.

The crystals proustite and tellurium are both absorbing and as there is no theory which satisfactorily treats absorbing crystals, it was decided to extend the theory to cover these cases. The crystal tellurium is also strongly gyrotropic and it was not known what effect this property would have on the phase matching condition. A theory was consequently developed for nonlinear interactions in gyrotropic media.

It should be mentioned that nonlinear optics is a comparatively new field in which a considerable amount of experimental and theoretical work is being done.

The theory of the optics of linear crystals has been studied extensively and is well described (Szivessy 1928). More recently the interaction of infinite plane waves has been considered in crystals which have a nonlinear constitutive relation between D and E (ABDP 1962), but which do not display optical activity or are lossy.

II LINEAR CRYSTAL OPTICS

This chapter is concerned with the propagation of electromagnetic radiation in linear media. Its purpose is to introduce a terminology which will serve for the rest of the thesis and to describe the simpler case of linear crystal optics before considering nonlinear propagation.

The history of the optics of linear crystals is long, comprehensive and is well documented (Szivessy 1928, Ram. and Ram. 1961). Consequently there is no need to discuss in detail the solving of the field equations here and it is only necessary to present the solutions of the field equations.

The propagation of electromagnetic radiation in crystals is only slightly more complicated than the more familiar case of propagation in isotropic media. In all respects the equations are of a more general form and all solutions must reduce to the isotropic ones when the tensor properties associated with the crystal become scalar or zero, depending on the tensor property. The effects observed in crystal optics can be explained in terms of two different theories, the elastic aether theory and Maxwell's theory. This duality of approach has led to many terminological difficulties. In this thesis the analysis is based upon Maxwell's electromagnetic theory and the emphasis throughout is on clarity and simplicity of approach.

2.1 The Field Equations and the Linear Constitutive Relations

The propagation of light in crystals both linear and nonlinear is governed, as in isotropic media, by Maxwell's equations; in c.g.s. units, for media with no free charge these are :

$$\nabla \times \underline{H} - \frac{1}{c} \frac{\partial \underline{D}}{\partial t} = 4\pi \underline{j} \quad \dots (2.1)$$

$$\nabla \times \underline{E} + \frac{1}{c} \frac{\partial \underline{B}}{\partial t} = 0 \quad \dots (2.2)$$

$$\nabla \cdot \underline{D} = 0 \quad \dots (2.3)$$

$$\nabla \cdot \underline{B} = 0 \quad \dots (2.4)$$

These equations are not sufficient as they describe wave propagation and it is necessary to introduce additional relationships which relate the field vectors \underline{B} and \underline{H} , \underline{D} and \underline{E} and \underline{j} and \underline{E} . These relationships are known as the constitutive relations and have always been known to be approximations. For absorbing, non-optically-active crystals which are electrically and magnetically anisotropic they are :

$$\underline{D} = \underline{\epsilon} \cdot \underline{E} \quad \dots (2.5)$$

$$\underline{B} = \underline{\mu} \cdot \underline{H} \quad \dots (2.6)$$

$$\underline{j} = \underline{\sigma} \cdot \underline{E} \quad \dots (2.7)$$

where $\underline{\epsilon}$, $\underline{\mu}$, $\underline{\sigma}$ are symmetric second-rank tensors of dielectric constant, magnetic permeability and conductivity respectively; all

the elements of these tensors are real. At optical frequencies the magnetic permeability tensor approaches a scalar value which in c.g.s. units is unity (B and W 1965).

The case of gyrotropic media is more complex. For infinite plane waves traversing non-absorbing, gyrotropic crystals the constitutive relation between \underline{D} and \underline{E} is :

$$\underline{D} = \underline{\epsilon} \cdot \underline{E} + i (\underline{G} \times \underline{E}) \quad \dots (2.8)$$

where \underline{G} is a vector known as the gyration vector and is derived from the relation:

$$\underline{G} = \underline{g} \cdot \underline{s} \quad \dots (2.9)$$

Here \underline{s} is a unit vector in the direction of propagation of the wave and \underline{g} is a tensor known as the gyration tensor and is characteristic of the medium; it is not necessarily symmetric, but the elements are real.

The more general case of absorbing, optically-active crystals requires the constitutive relation between \underline{D} and \underline{E} to be :

$$\underline{D} = \underline{\epsilon} \cdot \underline{E} + i \underline{G}' \times \underline{E} \quad \dots (2.10)$$

where again $\underline{G} = \underline{g} \cdot \underline{s}$. The tensor \underline{g} is now composed of a real part and an imaginary part neither of which is necessarily symmetric.

$$\underline{g} = \underline{h} + i \underline{k} \quad \dots (2.11)$$

Each of these tensors cause special effects and are not independent of one another. By making all of the tensors except one equal to zero or to a scalar, depending on the tensor, it is possible to see the effects each has.

The tensor $\tilde{\epsilon}$ has the effect that waves propagating through a crystal are linearly polarized and each linearly polarized component has a certain propagation constant which is a function of the angle of propagation of the wave with respect to the crystallographic axis. This tensor is also responsible for the fact that in general the two polarized components do not propagate their energy in the same direction. An example of the first effect is the interference pattern seen in the conoscopic photographs which are obtained when crystal slabs are illuminated with linearly polarised convergent light and are viewed through an analyser and an example of the second effect is the familiar effect of double images seen when an object is viewed through a calcite crystal.

The effects of the conductivity tensor are that the absorption of the waves is different for the characteristic polarizations and for different directions of propagation in the crystal. Usually the effect is linked with anisotropy of the dielectric constant and in this case the polarization states characteristic of a given direction are elliptical and in general they are not orthogonal. An example which shows that the absorption is different for different directions is given by the phenomena of pleochroism which enables tourmaline or Polaroid to be used as a polarizer. An example of the non-orthogonality of polarization states is the appearance of idiophanic fringes when a crystal is viewed in convergent light using only a polarizer or analyser.

The gyration tensor causes the characteristic polarization states to be circular and leads to the effect of optical rotation. This effect is caused by the incident linearly-polarized wave being decomposed into two circularly-polarized waves which pass through the gyrotropic media with different propagation constants and then interfere to produce linearly-polarized radiation. When the dielectric tensor does not behave as a scalar the situation becomes slightly more complicated. The characteristic polarizations become elliptical and are orthogonal. The eccentricity and orientation of the ellipses is a function of the direction of propagation as is the refractive index for each polarization state. The refractive indices experienced by the waves are not quite those obtained from the dielectric tensor, but are also dependent upon the gyration tensor. A familiar crystal which exhibits these effects is quartz. Here the polarization states for propagation down the optic axis are circular while the polarization states for propagation at right angles to it are linear; there are elliptically-polarized states for intermediate directions.

2.2 The Solution of the Field Equations for Infinite Plane Waves

Traversing Crystals

As has been mentioned earlier the solution of the field equations for the linear case is well documented (Szivessy 1928, Ram. and Ram. 1961) and consequently the final result presented here

discusses only the solution of the field equations for infinite plane waves of radian frequency ω .

Each of the field vectors E, D, H and B are of the form

$$\underline{A} = |A| \hat{a} \exp [i\omega(\frac{n'}{c} \underline{r} \cdot \underline{s} - t)] \dots (2.12)$$

where $|A|$ is the amplitude of the wave and is a constant, independent of distance and time; it will be seen that when we come to discuss nonlinear effects $|A|$ is no longer a constant.

The unit vector \hat{a} represents the state of polarization of the wave; this vector can be complex in the sense that it can be of the form $\hat{a} = \underline{b} + i \underline{c}$. The meaning of this is that the vector A is composed of the sum of two transverse sinusoidal vibrations with a constant phase relation between them and thus the propagating modes are of elliptical polarization. It is still possible to have an orthogonal polarization, \hat{a}' , to an elliptical polarization \hat{a} .
an example is left and right circularly polarized light

$$\hat{a}' = \underline{d} + i \underline{e} \quad (2.13)$$

The ellipticity and the orientation of the ellipse are given by solving the field equations.

In equation 2.12 n' is the complex refractive index associated with a given direction and is comprised of a real part which governs the phase velocity and an imaginary part which is related to the absorption coefficient.

r is a position vector, $\underline{r} = \hat{i}x + \hat{j}y + \hat{k}z$

s is a unit vector in the direction of propagation and t

represents time.

2.3 Solution of the Field Equations for other than Infinite Plane

Waves

The solutions which are found in the literature are mainly of the homogeneous plane wave type. It is also possible to solve for inhomogeneous plane waves, but it is difficult to solve for more general waves. The wave equation is, in general, not separable when there are more than two tensor properties which, when reduced to canonical form, do not have coincident principal axes. In any case the equations are only separable in one system of cartesian coordinates which means that the method of separation of variables is only of limited usefulness in practice.

The general method of solution is to meet a boundary condition by the addition of an infinite number of homogeneous and inhomogeneous plane waves travelling in all directions, then to allow each plane wave to propagate and find the effect at any point by the interference of the propagated waves.

It should be mentioned here that the method of superimposition is not valid in nonlinear media. The reason for solving for infinite plane waves in chapter 3 is not to form a basis on which to obtain a general solution, but to enable the only case which can be solved exactly to be considered. It should also be remarked that there is no solution which predicts correctly the second harmonic

power output when a crystal is illuminated with laser light and
there is significant conversion of fundamental to second harmonic.

III THE CONSTITUTIVE RELATION BETWEEN D AND E IN NONLINEAR MEDIA
AND THE TENSOR

No original work is presented in this chapter which is consequently kept as short as possible.

It has always been known that the linear constitutive relations $\underline{D} = \underline{\epsilon} \cdot \underline{E}$ and $\underline{B} = \underline{\mu} \cdot \underline{H}$ are approximations (Bloemberg 1964). The magnetic nonlinearity has been well investigated and is used in magnetic amplifiers while the non-linearity associated with ferromagnetic resonance has been used to generate second and higher harmonics in the microwave region of the spectrum. Until quite recently (1960) nonlinearities had not been observed at optical frequencies because the effects were very small. However the advent of the laser has meant that high power densities ($> 10^6$ watts/cm²) can be realised thus enabling the nonlinear effects to be demonstrated.

The nonlinear properties which will be considered in the rest of this thesis arise from the nonlinearity between D and E. The relation $\underline{B} = \underline{H}$ is taken to be valid at optical frequencies.

The relation between D and E is obtained by expanding the polarization as a power series in the electric field. For the pure electric dipole case we have:

$$\underline{P} = \underline{\chi}' \cdot \underline{E} + \underline{\chi}^2 \cdot \underline{EE} + \underline{\chi}^3 \cdot \underline{EEE} + \dots \dots \quad (3.1)$$

where the tensors $\underline{\chi}^n$ denote the linear and higher-order susceptibilities. It should be added that expansion 1 is not the most general one and that it would be better to use a multipole

expansion (Terhune 1964) then effects such as second harmonic generation in calcite could be explained.

In what follows the polarization will be considered to be composed of the linear susceptibility and the first nonlinear contribution then :

$$\underline{D} = \underline{\epsilon} \cdot \underline{E} + 4\pi \underline{\chi} : \underline{E} \underline{E} \quad \dots (3.2)$$

The nonlinear tensor $\underline{\chi}$ has certain symmetry conditions imposed on it by the material, just as has the linear dielectric tensor. The tensor $\underline{\chi}$ has the same symmetry as the piezoelectric tensor which means that all the terms are identically equal to zero whenever the medium has a centre of inversion. When the medium does not have this centre of inversion some of the elements are equal or zero in the crystals of higher symmetry. These equal and zero terms are to be found elsewhere and so need no further discussion.

The tensor $\underline{\chi}$ can be complex and this leads to phase shifts being introduced between the interacting waves which are not present when the tensor $\underline{\chi}$ is real (Bloembergen 1964).

IV WAVE PROPAGATION IN NONLINEAR CRYSTALS

4.1 Parametric Effects in Nonlinear, Nonabsorbing Anisotropic Media

The essential results presented in this section were first derived in (ABDP 1962); however the approach used is slightly different and forms the basis for the entirely original work in the next two sections on parametric effects in absorbing crystals and optically active crystals.

We start from Maxwell's equations and making the assumptions $\underline{B} = \underline{H}$ and $\underline{D} = \underline{\epsilon} \cdot \underline{E} + 4\pi \underline{P}_{NL}$ this gives:

$$\nabla \times \underline{E} = -\frac{1}{c} \frac{\partial \underline{H}}{\partial t} \quad \dots (4.1)$$

$$\nabla \times \underline{H} = \frac{1}{c} \frac{\partial}{\partial t} (\underline{\epsilon} \cdot \underline{E} + 4\pi \underline{P}_{NL}) \quad \dots (4.2)$$

We now take the curl of (1) and substitute from (2).

$$\nabla \times \nabla \times \underline{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} (\underline{\epsilon} \cdot \underline{E}) = -\frac{4\pi}{c^2} \frac{\partial^2 \underline{P}_{NL}}{\partial t^2} \quad \dots (4.3)$$

\underline{P}_{NL} is a source term of radian frequency ω . For simplicity we will consider all the waves involved to be monochromatic. Then the operation $\frac{\partial^2}{\partial t^2}$ is equivalent to multiplication by $-\omega^2$, so that

$$\nabla \times \nabla \times \underline{E} - \frac{\omega^2}{c^2} \underline{\epsilon} \cdot \underline{E} = 4\pi \frac{\omega^2}{c^2} \underline{P}_{NL} \quad \dots (4.4)$$

It should be noted that this is the usual linear wave equation with a source term.

We now consider the interaction of three infinite plane

waves travelling in the $+z$ direction, with propagation constants, k_1, k_2, k_3 and with radian frequencies $\omega_1, \omega_2, \omega_3$ respectively.

At ω_1

$$\underline{E}_1(z, t) = \hat{\underline{e}}_1 A_1(z) \exp[i(k_1 z - \omega_1 t)] \quad \dots (4.5)$$

$$\underline{P}_{1NL}(z, t) = \chi(\omega_1 = \omega_3 - \omega_2) : \underline{E}_3(z, t) \underline{E}_2^*(z, t) \dots (4.6)$$

At ω_2

$$\underline{E}_2(z, t) = \hat{\underline{e}}_2 A_2(z) \exp[i(k_2 z - \omega_2 t)] \quad \dots (4.7)$$

$$\underline{P}_{2NL}(z, t) = \chi(\omega_2 = \omega_3 - \omega_1) : \underline{E}_3(z, t) \underline{E}_1^*(z, t) \dots (4.8)$$

At ω_3

$$\underline{E}_3(z, t) = \hat{\underline{e}}_3 A_3(z) \exp[i(k_3 z - \omega_3 t)] \quad \dots (4.9)$$

$$\underline{P}_{3NL}(z, t) = \chi(\omega_3 = \omega_1 + \omega_2) : \underline{E}_1(z, t) \underline{E}_2(z, t) \dots (4.10)$$

Separating out each frequency term when substituted into (5) and then by scalar multiplication with the unit vector of \underline{E} we find

$$\hat{\underline{e}}_1 \cdot \nabla \times \nabla \times \underline{E}_1 - \left(\frac{\omega_1^2}{c^2} \right) \underline{\epsilon}(\omega_1) \cdot \underline{E}_1 = 4\pi \left(\frac{\omega_1^2}{c^2} \right) \chi(\omega_1) : \underline{E}_3 \underline{E}_2^* \dots (4.11)$$

$$\hat{\underline{e}}_2 \cdot \nabla \times \nabla \times \underline{E}_2 - \left(\frac{\omega_2^2}{c^2} \right) \underline{\epsilon}(\omega_2) \cdot \underline{E}_2 = 4\pi \left(\frac{\omega_2^2}{c^2} \right) \chi(\omega_2) : \underline{E}_3 \underline{E}_1^* \dots (4.12)$$

$$\hat{\underline{e}}_3 \cdot \nabla \times \nabla \times \underline{E}_3 - \left(\frac{\omega_3^2}{c^2} \right) \underline{\epsilon}(\omega_3) \cdot \underline{E}_3 = 4\pi \left(\frac{\omega_3^2}{c^2} \right) \chi(\omega_3) : \underline{E}_1 \underline{E}_2^* \dots (4.13)$$

In Appendix I an expression is derived for $\hat{\underline{e}} \cdot \nabla \times \nabla \times \underline{E}$ where \underline{E} represents a vector wave propagating in the z direction and $\hat{\underline{e}}$ is a unit vector of \underline{E} the result is:

$$\hat{e} \cdot \nabla \times \nabla \times \underline{E} = -\cos^2 \alpha \frac{d^2 \underline{E}}{dz^2} \quad \dots (4.14)$$

where $\alpha = \pi/2$ - the angle between the z direction and the direction of \underline{E} . In crystal optics α is the same as the angle between the wave normal and ray directions.

In Appendix II an expression is derived for $\hat{e} \cdot \underline{E} \cdot \underline{E}$ which is :

$$\hat{e} \cdot \underline{E} \cdot \underline{E} = n^2 \cos^2 \alpha |\underline{E}| \quad \dots (4.15)$$

where n is the phase refractive index.

Substituting (14) and (15) into (11), (12) and (13) yields equations of the form :

$$\frac{d^2 |\underline{E}|}{dz^2} + \frac{\omega^2 n^2}{c^2} |\underline{E}| = -\frac{4\pi \omega^2}{c^2 \cos^2 \alpha} \hat{e} \cdot \underline{P}_M \quad \dots (4.16)$$

Now

$$\frac{d^2}{dz^2} [A(z) \exp i(kz - \omega t)] = \exp i(kz - \omega t) \times \left[\frac{d^2 A}{dz^2} + 2ik \frac{dA}{dz} - k^2 A \right] \dots (4.17)$$

In physically realisable situations, the relative change in the amplitude per wavelength is small, since the nonlinear susceptibility is very small compared to the linear part. Thus terms in the second derivative of amplitude are negligible compared with the first derivatives, so (Bloem. 1964) :

$$\frac{d^2 A}{dz^2} \ll \frac{dA}{dz} \quad \dots (4.18)$$

After substitution and simplification we find :

$$\frac{dA_1}{dz} = \frac{i 2\pi \omega_1^2}{k_1 \cos \alpha_1 c^2} \hat{e}_1 \cdot \chi(\omega_1 = \omega_3 - \omega_2) \cdot \hat{e}_2 \hat{e}_3 A_3 A_2^* \exp(i \Delta k z) \quad (4.19)$$

where $\Delta k = k_3 - k_2 - k_1$.

From the fact that a photon at the pump frequency splits into one at the idler plus one at the signal frequency it is found that

$$K = \frac{2\pi}{c^2 \cos^2 \alpha_1} \hat{e}_1 \cdot \chi(\omega_1) : \hat{e}_2 \hat{e}_3 = \frac{2\pi}{c^2 \cos^2 \alpha_2} \hat{e}_2 \cdot \chi(\omega_2) : \hat{e}_1 \hat{e}_3 = \frac{2\pi}{c^2 \cos^2 \alpha_3} \hat{e}_3 \cdot \chi(\omega_3) : \hat{e}_1 \hat{e}_2 \quad (4.20)$$

Hence:

$$\frac{dA_1}{dz} = i \left(\frac{K \omega_1^2}{k_1} \right) A_3 A_2^* \exp(i \Delta k z) \quad \dots (4.21)$$

$$\frac{dA_2}{dz} = i \left(\frac{K \omega_2^2}{k_2} \right) A_3 A_1^* \exp(i \Delta k z) \quad \dots (4.22)$$

$$\frac{dA_3}{dz} = i \left(\frac{K \omega_3^2}{k_3} \right) A_1 A_2 \exp(-i \Delta k z) \quad \dots (4.23)$$

These equations will be called the parametric equations. They are a set of three nonlinear differential equations which can be solved exactly by the methods of (ABDP 1962) or approximately by assuming A_3 is independent of z .

4.2 Parametric Effects in Gyrotropic Crystals

The problem of parametric interactions in gyrotropic crystals has not been considered previously in the literature. This section is probably the most valuable part of the thesis and is not only of academic interest, but is also of great practical importance.

because all gyrotropic crystals lack a centre of inversion which means they can exhibit a first-order nonlinearity. Such potentially useful crystals as tellurium, cinnabar and lithium niobate are strongly gyrotropic.

In what follows the interaction of plane waves in a medium which has anisotropy of the dielectric tensor and is gyrotropic is considered. This situation, as will be seen, is somewhat similar to the case of parametric amplification in nonabsorbing crystals which possess anisotropy of the dielectric tensor only, but it is complicated by the fact that the polarizations of the interacting waves are no longer linearly polarized, but are, in general, elliptically polarized. In this chapter crystals which are non-absorbing are considered, so that the two characteristic polarization states are elliptical and orthogonal.

Derivation of the differential equations governing the amplitudes of the interacting waves as they pass through the nonlinear media

In what follows, the elliptical polarizations are represented by the addition of two mutually-orthogonal-linearly-polarized components with a constant phase relation between them.

From Maxwell's equations, as in chapter 1, for monochromatic radiation of radian frequency ω we obtain the relation:

$$\nabla \times \nabla \times \underline{E} = \frac{\omega^2}{c^2} \underline{D} \quad \dots (4.24)$$

The constitutive relation between \underline{D} and \underline{E} is taken as :

$$\underline{D} = \underline{\epsilon} \cdot \underline{E} + i \underline{G} \times \underline{E} + 4\pi \underline{P}_{NL} \quad \dots (4.25)$$

This relation is chosen because when the nonlinearity drops to zero the expression becomes $\underline{D} = \underline{\epsilon} \cdot \underline{E} + i \underline{G} \times \underline{E}$ which is the usual relation taken in linear crystal optics and when the vector \underline{G} is zero indicating no optical activity it becomes, $\underline{D} = \underline{\epsilon} \cdot \underline{E} + 4\pi \underline{P}_{NL}$ which is the expression taken when discussing nonlinear effects in nongyrotropic crystals.

Consequently $\nabla \times \nabla \times \underline{E} = \frac{\omega^2}{c^2} (\underline{\epsilon} \cdot \underline{E} + 4\pi \underline{P}_{NL} + i \underline{G} \times \underline{E}) \quad \dots (4.26)$

Equation 4.3 is now multiplied scalarly by $\hat{\underline{\epsilon}}$ and the results of Appendices I and II are utilized to obtain :

$$\frac{d^2 |E|}{dz^2} = \frac{\omega^2 n^2}{c^2} |E| + \frac{4\pi}{n^2 \cos^2 \alpha} \left(\frac{\omega^2}{c^2} \right) \hat{\underline{\epsilon}} \cdot \underline{P}_{NL} \quad \dots (4.27)$$

In the absence of any nonlinearities one solution of this equation represents a wave travelling in the positive z direction given by :

$$E(z) = A \exp(i k z) \quad \dots (4.28)$$

where A is an amplitude which is independent of z.

In the presence of nonlinearities we postulate the solution to be :

$$E(z) = A(z) \exp(i k z) \quad \dots (4.29)$$

$A(z)$ is governed by the differential equation :

$$\left(\frac{d^2 A}{dz^2} + 2ik \frac{dA}{dz} \right) \exp(i k z) = \frac{4\pi \omega^2}{n^2 \cos^2 \alpha} \frac{P_{NL}}{c^2} \cdot \hat{\underline{\epsilon}} \quad \dots (4.30)$$

as before the term $\frac{d^2A}{dz^2}$ is ignored in comparison with the term $2ik\frac{dA}{dz}$ which gives:

$$\frac{dA}{dz} = +i\frac{2\pi\omega^2}{kn^2c^2\cos\alpha} P_m \cdot \hat{e} \exp(-ikz) \quad \dots (4.31)$$

The nonlinear term is of interest and should be considered in slightly greater depth.

$$P_m \cdot \hat{e} = \hat{e}_1 \cdot \chi \cdot \hat{e}_2 \hat{e}_3 \quad \dots (4.32)$$

The unit vectors \hat{e}_j are complex and are of the form:

$$\hat{e}_j = \hat{a}_j + i \hat{b}_j \quad \dots (4.33)$$

where $j = 1, 2, 3$

then $\hat{e}_1 \cdot \chi \cdot \hat{e}_2 \hat{e}_3 = (\hat{a}_1 + i \hat{b}_1) \cdot \chi \cdot [\hat{a}_2 \hat{a}_3 + \hat{b}_2 \hat{b}_3 + i(\hat{b}_2 \hat{a}_3 + \hat{a}_2 \hat{b}_3)] \quad (4.34)$

$$= \hat{a}_1 \cdot \chi \cdot (\hat{a}_2 \hat{a}_3 - \hat{b}_2 \hat{b}_3) - \hat{b}_1 \cdot \chi \cdot (\hat{b}_2 \hat{a}_3 + \hat{a}_2 \hat{b}_3) \dots (4.35) \\ + i[\hat{b}_1 \cdot \chi \cdot (\hat{a}_2 \hat{a}_3 - \hat{b}_2 \hat{b}_3) + \hat{a}_1 \cdot \chi \cdot (\hat{b}_2 \hat{a}_3 + \hat{a}_2 \hat{b}_3)]$$

The tensor χ is known and the vectors a_i and b_i are also known; it is thus possible to calculate the coupling term. It should also be remarked that it is necessary to use the approach developed in this chapter to determine the elements of the nonlinear tensor χ . It will be appreciated that the calculation of the coupling term is lengthy and somewhat involved.

There are two cases of parametric interaction which must be dealt with. They are the situation of second harmonic generation and the case of parametric amplification.

Second-harmonic generation

Some work has appeared in the literature on SHG in gyrotropic crystals and it is appropriate at this point to mention what has been done. The only theoretical work consists of two papers published by Rabin and Bey (Rabin and Bey 1967, 1967). These papers are concerned with the effects of optical activity on phase matching and they tackle the problem by two different approaches. The first of these is similar to Franken and Ward method (Franken and Ward, 1963) where the fundamental is considered to establish a phased three-dimensional array of dipoles at the second-harmonic frequency, which then radiate and second harmonic radiation is emitted. This method is only approximate because no allowance can be made for the pump wave becoming weaker due to conversion to second harmonic. To overcome this deficiency Rabin and Bey published a second paper which was of a coupled-wave approach as is the work described here.

The two papers were concerned only with waves which are circularly polarized. The results given here are much more general and apply equally to elliptically polarized waves as well as to the special case of circular polarization. When the interacting waves are allowed to become circularly polarized the growth equations reduce to those of Rabin and Bey. It should be mentioned, however, that Rabin and Bey have not used quite the same definition of k , the propagation constant, as I have. They have taken k as that part of the propagation constant which arises from the dielectric

tensor and have added to this another part, α , which comes from the gyration tensor. In what is presented here these two terms are added, or subtracted, and means this work falls in line with the treatment given by texts on linear crystal optics (Born and Wolf, 1965).

The equations governing the growth of second harmonic

For second-harmonic generation the fundamental establishes a nonlinear polarization at twice the optical frequency

$$P_{NL_2} = \tilde{\chi}(\omega_1, \omega_1, \omega_1) : \underline{E} \underline{E} \quad \dots (4.36)$$

There is also a mixing between the second harmonic and the fundamental which yields:

$$P_{NL_1} = \tilde{\chi}(\omega_1, \omega_1, \omega_1) : \underline{E}_2 \underline{E}_1^* \quad \dots (4.37)$$

The equations governing the amplitudes are :

$$\frac{dA_2}{dy} = -i \frac{\omega_1^2 2\pi}{c^2 k n_1^2 \cos^2 \alpha_2} \hat{\underline{e}}_2 \cdot \tilde{\chi}(\omega_1, \omega_1, \omega_1) : \hat{\underline{e}}_1 \hat{\underline{e}}_1^* A_1 A_1^* \exp(-i\Delta k_2) \quad \dots (4.38)$$

$$\frac{dA_1}{dy} = -i \frac{\omega_1^2 2\pi}{c^2 k n_1^2 \cos^2 \alpha_1} \hat{\underline{e}}_1 \cdot \tilde{\chi}(\omega_1, \omega_1, \omega_1) : \hat{\underline{e}}_2 \hat{\underline{e}}_2^* A_2 A_2^* \exp(i\Delta k_1) \quad \dots (4.39)$$

here $\Delta k = k_2 - 2k_1 \quad \dots (4.40)$

These equations are of exactly the same form as the equations governing second-harmonic generation in nongyrotropic media. The differences are chiefly in the coupling terms $\hat{\underline{e}} \cdot \tilde{\chi} : \hat{\underline{e}} \hat{\underline{e}}^*$ and in the unit vectors $\hat{\underline{e}}$.

Parametric amplification in gyrotropic media

Here there are three waves interacting; the pump, the signal and the idler.

The pump and signal mix to produce a nonlinear polarization at the idler frequency. In the following the subscript '1' refers to the signal, '2' to the idler and '3' to the pump wave. Thus

$$P_{NL} = \chi(\omega_3, \omega_1, \omega_2) : \hat{e}_3 \hat{e}_2^* A_3 A_2 \exp[i(\mathbf{k}_3 - \mathbf{k}_2) \cdot \mathbf{r}] \quad \dots (4.41)$$

The growth equation for the signal is thus:

$$\frac{dA_1}{dz} = -i \frac{2\pi \omega_1^2}{k_1 n_1^2 \cos^2 \alpha_1 C^2} \hat{e}_1 \cdot \chi : \hat{e}_3 \hat{e}_2^* A_3 A_2 \exp(i\Delta k_3) \dots (4.42)$$

where $\Delta k = k_3 - k_2 - k_1$

Similarly the signal and pump mix to produce a nonlinear polarization at the signal frequency and the growth equation is:

$$\frac{dA_2}{dz} = -i \frac{2\pi \omega_2^2}{k_2 n_2^2 \cos^2 \alpha_2 C^2} \hat{e}_2 \cdot \chi : \hat{e}_3 \hat{e}_1^* A_3 A_1 \exp(i\Delta k_3) \dots (4.43)$$

The signal and idler also mix to produce radiation at the pump frequency

$$\frac{dA_3}{dz} = -i \frac{2\pi \omega_3^2}{k_3 n_3^2 \cos^2 \alpha_3 C^2} \hat{e}_3 \cdot \chi : \hat{e}_1 \hat{e}_2^* A_1 A_2 \exp(-i\Delta k_3) \dots (4.44)$$

The equations above govern the amplitudes of the waves as they pass through the crystal. It is possible, however, to find a useful relationship by requiring the number of photons at the pump frequency plus the number at the signal frequency to be constant, and the number of photons at the idler frequency plus the number at the

pump frequency to be another constant independent of z . This is because a pump photon splits into one signal plus one idler photon.

Then

$$\frac{d}{dz} \left(\frac{A_1^* A_1}{\omega_1} \right) = \frac{d}{dz} \left(\frac{A_2^* A_2}{\omega_2} \right) = - \frac{d}{dz} \left(\frac{A_3^* A_3}{\omega_3} \right) \quad \dots (4.45)$$

From equations (42) to (45) a relation can be found between the terms $\hat{e}_2 \cdot \chi \cdot \hat{e}_3 \hat{e}_1^*$. Equation (43) is divided by (42) and after simplification and manipulation, as in section 1 of this chapter, it is found that to be consistent with equation (45) we require

$$\frac{\hat{e}_2 \cdot \chi \cdot \hat{e}_3 \hat{e}_1^*}{\cos^2 \alpha_2} = \frac{\hat{e}_1 \cdot \chi \cdot \hat{e}_3 \hat{e}_2^*}{\cos^2 \alpha_1} \quad \dots (4.46)$$

and from equation (43) and the complex conjugate of equation (44)

$$\frac{\hat{e}_2 \cdot \chi \cdot \hat{e}_3 \hat{e}_1^*}{\cos^2 \alpha_2} = \frac{\hat{e}_3 \cdot \chi \cdot \hat{e}_1 \hat{e}_2^*}{\cos^2 \alpha_3} \quad \dots (4.47)$$

Hence

$$\frac{\hat{e}_1 \cdot \chi \cdot \hat{e}_3 \hat{e}_2^*}{\cos^2 \alpha_1} = \frac{\hat{e}_2 \cdot \chi \cdot \hat{e}_3 \hat{e}_1^*}{\cos^2 \alpha_2} = \frac{\hat{e}_3 \cdot \chi \cdot \hat{e}_1 \hat{e}_2^*}{\cos^2 \alpha_3} = K \quad \dots (4.48)$$

Thus the parametric equations are :

$$\frac{dA_1}{dz} = -\frac{i 2 \pi \omega_1^2}{k_1 n_1^2 c^2} K A_3 A_1^* \exp(i \Delta k z) \quad \dots (4.49)$$

$$\frac{dA_2}{dz} = -\frac{i 2 \pi \omega_2^2}{k_2 n_2^2 c^2} K A_3 A_2^* \exp(i \Delta k z) \quad \dots (4.50)$$

$$\frac{dA_3}{dz} = -\frac{i 2 \pi \omega_3^2}{k_3 n_3^2 c^2} K A_1 A_2 \exp(-i \Delta k z) \quad \dots (4.51)$$

This set of three nonlinear differential equations can be solved exactly by the methods of (ABDP, 1962) or by the small gain theory whereby A_3 is regarded as being independent of z and then the equations become linear and are soluble by the usual methods for simultaneous linear differential equations.

The effect of the medium being gyrotropic is that the nonlinear coupling term $\hat{e} \cdot \chi \cdot \hat{e} \hat{e}$ is no longer the same as for the corresponding nongyrotropic medium and this term is also complex which means that there will be effects produced which are akin to those produced by a complex χ tensor - that is extra phase shifts will be produced in the waves as mentioned in chapter 2. In all other ways however the waves behave like the more familiar linearly polarized ones.

4.3 Parametric Effects in Nonlinear, Absorbing, Anisotropic Media

The following derivation of the parametric growth equations is not to be found elsewhere. There are two things which justify the inclusion of this work in the thesis. The first is that it extends the theory of the previous sub-section and enables real crystals, which have absorption at the interacting frequencies, to be considered. This is of relevance to the experimental work being undertaken in the group because both the tellurium and proustite crystals are absorbing. The second is that a point arises which has not been mentioned elsewhere.

When an experiment is performed to measure nonlinear coefficients the polarizations are taken to be linear, and assuming this the unknown nonlinear coefficients can be determined. In fact, from the theory of linear crystal optics, the polarizations are known to be elliptical and allowance should be made for this ellipticity when calculating the nonlinear coefficients. The theory presented here allows the nonlinear coefficients to be determined correctly. More is said about this latter point when gyrotropic crystals are considered.

The effects of absorption are introduced by assuming the medium to have a finite conductivity at the frequencies concerned. This property is direction dependent and is formulated using the conductivity tensor, which need not have the same principal directions as the dielectric tensor. The derivation given here is general and applies to all nonlinear absorbing crystals, both uniaxial and biaxial which are not gyrotropic.

As in the last section we start with Maxwell's equations and take $\underline{B} = \underline{H}$. The interacting waves are assumed to be monochromatic and of radian frequency ω . In this case the operation $\frac{\partial}{\partial t}$ is equivalent to multiplication by $-i\omega$ giving:

$$\nabla \times \underline{E} = \frac{i\omega}{c} \underline{H} \quad \dots (4.52)$$

$$\nabla \times \underline{H} = \frac{4\pi\sigma}{c} \underline{E} - \frac{i\omega}{c} \underline{D} \quad \dots (4.53)$$

The curl of equation (52) is taken and substituted into equation (53) as before, and followed by scalar multiplication by $\hat{\underline{e}}$ which

is the complex unit vector of \underline{E} , and determines the ellipticity of its polarization. We have :

$$\hat{\underline{e}} \cdot \nabla \times \nabla \times \underline{E} - \frac{\omega^2}{c^2} \hat{\underline{e}} \cdot \left(\underline{\epsilon} \cdot \underline{E} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E} \right) = 4\pi \frac{\omega^2}{c^2} \hat{\underline{e}} \cdot \underline{P}_{NL} \dots (4.54)$$

As before (Appendix I) $\hat{\underline{e}} \cdot \nabla \times \nabla \times \underline{E} = -\cos^2 \alpha \frac{d^2 |E|}{dz^2}$

and in Appendix III it is shown that

$$\hat{\underline{e}} \cdot \left(\underline{\epsilon} \cdot \underline{E} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E} \right) = n' \cos^2 \alpha |E|$$

where n' is the complex refractive index and α is the angle between the Poynting vector and the wave normal. After making these substitutions equation 4.54 becomes

$$\frac{d^2 |E|}{dz^2} + \frac{\omega^2 n'^2}{c^2} |E| = - \frac{4\pi}{\cos^2 \alpha} \frac{\omega^2}{c^2} \hat{\underline{e}} \cdot \underline{P}_{NL} \dots (4.55)$$

Now $|E|$ is complex and is of the form :

$$|E| = B(z) \exp(i n' \frac{\omega}{c} z) \dots (4.56)$$

$$= B(z) \exp(-\beta z) \exp(i k z) \dots (4.57)$$

$$= A(z) \exp(i k z) \dots (4.58)$$

k is the propagation constant and β is the imaginary part of the complex propagation constant; in fact it is the amplitude absorption coefficient when there are no nonlinear interactions, i.e. at small signals.

Now $\frac{d^2 |E|}{dz^2} = \exp(i n' \frac{\omega}{c} z) \times \left(\frac{d^2 B}{dz^2} + 2i n' \frac{\omega}{c} \frac{dB}{dz} - n'^2 \frac{\omega^2}{c^2} B \right) \dots (4.59)$

This is then substituted into equation (57) and ignoring $\frac{\partial^2 B}{\partial z^2}$ compared with $2i n' \frac{\omega}{c} \frac{dB}{dz}$, as before, we obtain :

$$\frac{wn'}{c} \frac{dB}{dz} = \frac{i 2\pi \omega^2}{\cos^2 \alpha} \hat{e} \cdot P_{NL} \exp(-i n' \frac{\omega}{c} z) \quad \dots (4.60)$$

Now $\frac{wn'}{c} = k_x + i\beta$ and k is $\gg \beta$ thus in equation (60) we set $\frac{n' \omega}{c} = k_x$ on LHS of equation (60) and obtain:

$$\frac{dB}{dz} = i \frac{2\pi \omega^2}{k_x c^2 \cos^2 \alpha} \hat{e} \cdot P_{NL} \exp(-i k_x z) \exp(\beta z) \quad \dots (4.61)$$

Now from equations (57) and (58)

$$A(z) = B(z) \exp(-\beta z) \quad \dots (4.62)$$

so substituting from (62) into (61) gives

$$\frac{dA}{dz} + \beta A = i \frac{2\pi \omega^2}{k_x c^2 \cos^2 \alpha} \hat{e} \cdot P_{NL} \exp(-i k_x z) \quad \dots (4.63)$$

There are three equations like (63) governing the amplitudes of the waves:

$$\frac{dA_1}{dz} + \beta_1 A_1 = i \frac{2\pi \omega_1^2}{k_1 c^2 \cos^2 \alpha_1} \hat{e}_1 \cdot \hat{\chi} : \hat{e}_3 \hat{e}_2^* A_3 A_2^* \exp(i k_1 z). \quad (4.64)$$

$$\frac{dA_2}{dz} + \beta_2 A_2 = i \frac{2\pi \omega_2^2}{k_2 c^2 \cos^2 \alpha_2} \hat{e}_2 \cdot \hat{\chi} : \hat{e}_3 \hat{e}_1^* A_3 A_1^* \exp(i k_2 z) \quad (4.65)$$

$$\frac{dA_3}{dz} + \beta_3 A_3 = i \frac{2\pi \omega_3^2}{k_3 c^2 \cos^2 \alpha_3} \hat{e}_3 \cdot \hat{\chi} : \hat{e}_2 \hat{e}_1^* A_1 A_2^* \exp(i k_3 z) \quad (4.66)$$

Equations (64) to (66) are the parametric equations when there is absorption present. It will be noticed that these reduce to the parametric equations for nonabsorbing media when the quantities $\beta_1, \beta_2, \beta_3$ become zero.

These equations are more difficult to solve than those which arise for the lossless case. It is no longer possible to find an expression relating the terms $\underline{\underline{\epsilon}} \cdot \underline{\underline{\chi}} : \underline{\underline{\epsilon}} \underline{\underline{\epsilon}}$ for the equations (64) to (66). When these are obtained from experiment the quantity should be written as:

$$(\underline{a}_1 + i\underline{b}_1) \cdot \underline{\underline{\chi}} : [\underline{a}_2 \underline{a}_3 + \underline{b}_2 \underline{b}_3 + i(\underline{a}_1 \underline{b}_3 - \underline{b}_1 \underline{a}_3)] \quad \dots (4.67)$$

The quantities \underline{a} , \underline{b} decide the ellipticity of the waves and can be determined experimentally. The tensor elements of $\underline{\underline{\chi}}$ can be obtained.

It should be emphasised that this is not just of academic interest. The propagating modes are significantly different from linear and they are circular in certain directions known as the singular axes.

CHAPTER V SOLUTIONS OF THE PARAMETRIC EQUATIONS

1. Introduction

The parametric equations are derived in chapter 3. Some solutions of these equations are discussed in this chapter. The solution is of the small-signal type in which the depletion of the pump wave is neglected. The absorption at the pump frequency is also taken to be zero. The solutions are applicable to absorbing, optically-active and nonabsorbing crystals.

2. Solution of the equations

The relevant parametric equations are re-stated from chapter 3:

$$\frac{dA_1}{dz} = -\beta_1 A_1 + i c_1 A_2^* \exp(+i\Delta k z) \quad \dots (5.1)$$

$$\frac{dA_2}{dz} = -\beta_2 A_2 + i c_2 A_1^* \exp(+i\Delta k z) \quad \dots (5.2)$$

where $c_1 = \frac{2\pi w_1^2}{k_1 c^2 \cos^2 \alpha} \hat{e}_1 \cdot \hat{\chi} \cdot \hat{e}_2^* \hat{e}_2 A_3$... (5.3)

and $c_2 = \frac{2\pi w_2^2}{k_2 c^2 \cos^2 \alpha} \hat{e}_2 \cdot \hat{\chi} \cdot \hat{e}_3^* \hat{e}_3 A_3$... (5.4)

Now $\frac{dA_1}{dz} + \beta_1 A_1 = \exp(-\beta_1 z) \frac{d}{dz} (A_1 \exp \beta_1 z) \quad \dots (5.5)$

and $\frac{dA_2}{dz} + \beta_2 A_2 = \exp(-\beta_2 z) \frac{d}{dz} (A_2 \exp \beta_2 z) \quad \dots (5.6)$

Thus $\frac{d}{dz} (A_1 \exp \beta_1 z) = +i c_1 A_2^* \exp((\beta_1 + i\Delta k)z) \dots (5.7)$

and $\frac{d}{dz} (A_2 \exp \beta_2 z) = +i c_2 A_1^* \exp((\beta_2 + i\Delta k)z) \dots (5.8)$

The complex conjugate of (8) is :

$$\frac{d}{dz} (A_2^* \exp \beta_2 z) = -i C_2^* A_1 \exp ((\beta_2 - i \Delta k) z) \dots (5.9)$$

from (7) $A_2^* \exp \beta_2 z = -\frac{i}{C_1} \exp ((-i \Delta k - \beta_1 + \beta_2) z) \frac{d}{dz} (A_1 \exp \beta_1 z) \dots (5.10)$

from (9) $A_1 \exp \beta_1 z = +\frac{i}{C_1^*} \exp ((-i \Delta k - \beta_1 + \beta_2) z) \frac{d}{dz} (A_2^* \exp \beta_2 z) \dots (5.11)$

Now put $\rho = i \Delta k - \beta_1 + \beta_2 \dots (5.12)$

Then differentiating (10) w.r.t. z

$$\frac{d}{dz} (A_2^* \exp \beta_2 z) = \frac{i}{C_1^*} \exp (\rho z) \left\{ \frac{d^2}{dz^2} (A_1 \exp \beta_1 z) + \rho \frac{d}{dz} (A_1 \exp \beta_1 z) \right\} \dots (5.13)$$

Similarly differentiating (11) w.r.t. z gives

$$\frac{d}{dz} (A_1 \exp \beta_1 z) = -\frac{i}{C_1^*} \exp (-\rho z) \left\{ \frac{d^2}{dz^2} (A_2^* \exp \beta_2 z) - \rho \frac{d}{dz} (A_2^* \exp \beta_2 z) \right\} \dots (5.14)$$

Now substitute from (14) for $\frac{d}{dz} (A_1(z) \exp \beta_1 z)$ into (7)

$$-\frac{i}{C_1^*} \exp (-\rho z) \left\{ \frac{d^2}{dz^2} (A_2^* \exp \beta_2 z) - \rho \frac{d}{dz} (A_2^* \exp \beta_2 z) \right\} = i C_1^* A_2^* \exp (\beta_1 - i \Delta k z) \dots (5.15)$$

Simplification gives

$$\frac{d^2 A_2^* \exp \beta_2 z}{dz^2} - \rho \frac{d}{dz} (A_2^* \exp \beta_2 z) - C_1^* A_2^* \exp \beta_2 z = 0 \dots (5.16)$$

Substituting for $\frac{d}{dz} (A_2^* (z) \exp \beta_2 z)$ from (13) into (9) gives after re-arrangement:

$$\frac{d^2}{dz^2} (A_1 \exp \beta_1 z) + \rho \frac{d}{dz} (A_1 \exp \beta_1 z) - C_1 C_2^* (A_1 \exp \beta_1 z) = 0 \dots (5.17)$$

Equations (16) and (17) are equations which control the magnitudes and phases of the amplitudes. These equations will now be solved subject to boundary conditions :

$$A_2^*(0) = A_2^* \quad \dots (5.18)$$

$$A_1(0) = A_1^* \quad \dots (5.19)$$

$\frac{dA_1}{dz}$ and $\frac{dA_2^*}{dz}$ at $z = 0$ are obtained from equations (7) and (9). These boundary conditions represent a small signal A_1^* and a small idler A_2^* present at $z = 0$.

3. Applications of Boundary Conditions

The solution of (16) is :

$$A_2^* \exp \beta_1 z = \exp \left(\frac{P_1^2}{2} \right) \{ H \exp(\alpha z) + J \exp(-\alpha z) \} \dots (5.20)$$

and the solution of (17) is :

$$A_1 \exp \beta_1 z = \exp \left(- \frac{P_1^2}{2} \right) \{ F \exp(\alpha z) + G \exp(-\alpha z) \} \dots (5.21)$$

where

$$\alpha = \sqrt{\frac{P^2 + 4 \omega C_1 C_2^*}{2}}$$

We now determine F, G, H, J from the boundary conditions at $z = 0$ which are obtained from equations (7), (9), (18) and (19).

$$G = \frac{i C_1 A_2^*}{2\alpha} - \frac{\rho A_1^*}{4\alpha} + \frac{A_1^*}{2} \quad \dots (5.22)$$

$$F = -\frac{i C_1 A_1^*}{2\alpha} + \frac{\rho A_1^*}{4\alpha} + \frac{A_1^*}{2} \quad \dots (5.23)$$

$$J = -\frac{i C_2^* A_1^*}{2\alpha} + \frac{\rho A_2^*}{4\alpha} + \frac{A_2^*}{2} \quad \dots (5.24)$$

$$H = \frac{i C_2^* A_1^*}{2\alpha} - \frac{\rho A_2^*}{4\alpha} + \frac{A_2^*}{2} \quad \dots (5.25)$$

From (20), (21), (23), (24) and (25)

$$A_i \exp \beta_i z = \exp \left(-\frac{p z}{2} \right) \left\{ -\frac{i c_1 A_i^0}{\alpha} \sinh \alpha z + \frac{p A_i^0 \cosh \alpha z}{2\alpha} + A_i^0 \cosh \alpha z \right\} \dots (5.26)$$

$$A_i^0 \exp \beta_i z = \exp \left(\frac{p z}{2} \right) \left\{ \frac{i c_2^* A_i^0}{\alpha} \sinh \alpha z - \frac{p A_i^0 \cosh \alpha z}{2\alpha} + A_i^0 \sinh \alpha z \right\} \dots (5.27)$$

Equations (26) and (27) are in a more convenient form if written:

$$A_i(z) = A_i^0 M(z) + A_i^0 N(z) \dots (5.28)$$

$$A_i^0(z) = A_i^0 P(z) + A_i^0 Q(z) \dots (5.29)$$

where

$$M(z) = \exp \left(-\frac{\beta_1 + \beta_2 z}{2} \right) \exp \left(-\frac{i \Delta k z}{2} \right) \left(\cosh \alpha z + \frac{p}{2\alpha} \sinh \alpha z \right) \dots (5.30)$$

$$N(z) = \exp \left(-\frac{\beta_1 + \beta_2 z}{2} \right) \exp \left(-\frac{i \Delta k z}{2} \right) \left\{ -\frac{i c_1}{\alpha} \sinh \alpha z \right\} \dots (5.31)$$

$$P(z) = \exp \left(-\frac{\beta_1 + \beta_2 z}{2} \right) \exp \left(\frac{i \Delta k z}{2} \right) \left\{ \frac{i c_2^*}{\alpha} \sinh \alpha z \right\} \dots (5.32)$$

$$Q(z) = \exp \left(-\frac{\beta_1 + \beta_2 z}{2} \right) \exp \left(\frac{i \Delta k z}{2} \right) \left\{ \cosh \alpha z - \frac{p}{2\alpha} \sinh \alpha z \right\} \dots (5.33)$$

$$\alpha = \frac{\sqrt{p^2 + 4c_1 c_2}}{2} \dots (5.34)$$

$$p = i \Delta k - \beta_1 + \beta_2 \dots (5.35)$$

There is a relevant special case of these equations when the absorption at both the signal and idler frequencies are zero.

The equations are then simplified to :

$$A_i(z) = A_i^0 R(z) + A_i^0 S(z) \dots (5.36)$$

$$A_i^0(z) = A_i^0 T(z) + A_i^0 U(z) \dots (5.37)$$

where

$$R(z) = \exp\left(-\frac{i\Delta k z}{2}\right) \left\{ \cosh \alpha z + \frac{i\Delta k}{2\alpha} \sinh \alpha z \right\} \dots (5.38)$$

$$S(z) = \exp\left(-\frac{i\Delta k z}{2}\right) \left\{ -\frac{i c_1}{\alpha} \sinh \alpha z \right\} \dots (5.39)$$

$$T(z) = \exp\left(\frac{i\Delta k z}{2}\right) \left\{ \frac{i c_2^*}{\alpha} \sinh \alpha z \right\} \dots (5.40)$$

$$U(z) = \exp\left(\frac{i\Delta k z}{2}\right) \left\{ \cosh \alpha z - \frac{i\Delta k}{2\alpha} \sinh \alpha z \right\} \dots (5.41)$$

$$\alpha = \frac{\sqrt{4c_1 c_2 - \Delta k^2}}{2} \dots (5.42)$$

4. Parametric Amplification with No Absorption

This has been discussed elsewhere (Yariv 1967) consequently this section is very brief. In this case there is a signal A_1 at $z = 0$ and the idler at $z = 0$ is zero.

Hence from equations (36) to (42)

$$A_1(z) = A_1^0 \exp\left(-\frac{i\Delta k z}{2}\right) \left\{ \cosh \alpha z + i \frac{\Delta k}{2\alpha} \sinh \alpha z \right\} \dots (5.43)$$

and

$$A_2^*(z) = A_1^0 \exp\left(\frac{i\Delta k z}{2}\right) \left\{ \frac{i c_2^*}{\alpha} \sinh \alpha z \right\} \dots (5.44)$$

The amplitude of the signal wave entering the amplifying region is A_1^0 and that which leave is $A_1(\ell)$ and $A_2^*(\ell)$ where ℓ is the length of the nonlinear medium.

The power gain for the signal is thus :

$$\frac{A_s(\ell) A_i(\ell)}{A_s^0 A_i^0} = \left(\frac{i \Delta k \sinh \alpha z + \cosh \alpha z}{2\alpha} \right) \left(\frac{-i \Delta k \sinh \alpha^* z + \cosh \alpha^* z}{2\alpha} \right) \dots \quad (5.45)$$

$$= \frac{\Delta k^2}{4\alpha^2} \sinh \alpha z \sinh \alpha^* z + i \frac{\Delta k}{2} \left(\frac{\sinh \alpha z \cosh \alpha^* z - \cosh \alpha z \sinh \alpha^* z}{\alpha} \right) \dots \quad (5.46)$$

Then power gain =

$$\frac{I_s(\ell)}{I_s(0)} = \cosh((\alpha + \alpha^*)\ell) \left(\frac{1}{2} + \frac{\Delta k^2}{8\alpha^2} \right) + \cosh((\alpha - \alpha^*)\ell) \left(\frac{1}{2} - \frac{\Delta k^2}{8\alpha^2} \right) + \sinh((\alpha + \alpha^*)\ell) \left(\frac{i \Delta k}{4\alpha} - \frac{i \Delta k}{4\alpha} \right) + \sinh((\alpha - \alpha^*)\ell) \left(\frac{i \Delta k}{4\alpha} - \frac{i \Delta k}{4\alpha} \right) \dots \quad (5.47)$$

There are two cases to be considered.

Case 1 when $\Delta k^2 \leq 4c_1 c_2$

$$\text{Here } \alpha = \sqrt{4c_1 c_2 - \Delta k^2} \text{ is real} \dots \quad (5.48)$$

$$\text{Then } \alpha + \alpha^* = 2\alpha \dots \quad (5.49)$$

$$\text{and } \alpha - \alpha^* = 0 \dots \quad (5.50)$$

$$\text{Then } \frac{I_s(\ell)}{I_s(0)} = \cosh 2\alpha \ell \left(\frac{1}{2} + \frac{\Delta k^2}{8\alpha^2} \right) + \left(\frac{1}{2} - \frac{\Delta k^2}{8\alpha^2} \right) \dots \quad (5.51)$$

Case 2 $\Delta k^2 \geq 4c_1 c_2$

$$\text{Here } \alpha = \sqrt{4c_1 c_2 - \Delta k^2} \text{ is pure imaginary} \dots \quad (5.52)$$

$$\text{Then } \alpha + \alpha^* = 0 \dots \quad (5.53)$$

$$\text{and } \alpha - \alpha^* = 2i|\alpha| \dots \quad (5.54)$$

$$\frac{I_s(\ell)}{I_s(0)} = \cos(2|\alpha|\ell) \left(\frac{1}{2} + \frac{\Delta k^2}{8|\alpha|^2} \right) + \left(\frac{1}{2} - \frac{\Delta k^2}{8|\alpha|^2} \right) \dots \quad (5.55)$$

6. Parametric Amplifier with Absorption

Absorption is present, A_2^{*0} is zero and we require to find the power gain of the amplifier which is :

$$\frac{I_s(l)}{I_s(0)} = \frac{A_s(l)^* A_s(l)}{A_s(0)^* A_s(0)} \quad \dots (5.56)$$

Now from equations (28) to (35)

$$A_s(l) = A_s^0 \exp(-(\beta_1 + \beta_2)l) \exp\left(-\frac{i\Delta k l}{2} \frac{P \sinh \alpha l}{\alpha^2 + \cosh \alpha l} \dots\right) \quad (5.57)$$

hence

$$\frac{A_s(l)^* A_s(l)}{A_s^0 A_s^0} = \exp(-(\beta_1 + \beta_2)l) \left\{ \frac{P^* P}{4\alpha^2} \sinh \alpha l \cosh \alpha l + \frac{P \sinh \alpha l \cosh \alpha l}{2\alpha} \right\} \dots (5.58)$$

Simplifying

$$\frac{I_s(l)}{I_s(0)} = \exp(-(\beta_1 + \beta_2)l) \left\{ \cosh((\alpha + \alpha^*)l) \left(\frac{P^* P}{8\alpha^2} + \frac{1}{2} \right) + \cosh((\alpha - \alpha^*)l) \left\{ \frac{1}{2} - \frac{P^* P}{8\alpha^2 \alpha} \right\} \right. \\ \left. + \sinh((\alpha + \alpha^*)l) \left(\frac{P}{4\alpha} + \frac{P^*}{4\alpha^*} \right) + \sinh((\alpha - \alpha^*)l) \left(\frac{P}{4\alpha} - \frac{P^*}{4\alpha^*} \right) \right\} \quad (5.59)$$

Also $A_s(l)^* A_s(l) = A_s^0 A_s^0 P(l) P(l) \quad \dots (5.60)$

$$= A_s^0 A_s^0 \exp(-(\beta_1 + \beta_2)l) \left(\frac{c_1^* c_2}{\alpha^2} \sinh \alpha l \cosh \alpha^* l \right) \dots (5.61)$$

$$= A_s^0 A_s^0 \exp(-(\beta_1 + \beta_2)l) \frac{c_1^* c_2}{2\alpha} \left\{ \cosh((\alpha + \alpha^*)l) - \cosh((\alpha - \alpha^*)l) \right\} \dots (5.62)$$

Thus

$$\frac{I_s(l)}{I_s(0)} = \frac{n_1 w_1 c_1^* c_2}{n_2 w_2 2\alpha^2 \alpha} \exp(-(\beta_1 + \beta_2)l) \left\{ \cosh((\alpha + \alpha^*)l) - \cosh((\alpha - \alpha^*)l) \right\} \dots (5.63)$$

Equations (59) and (63) are interesting because they show that the effects of absorption are similar to those of the phase mismatch

Δk becoming complex. It is in fact related closely to the number p by equation (35). The quantity α is complex and the equations cannot be put in the simple form of (51) and (55).

The whole situation is much more complicated than the familiar lossless case and to clarify the situation it is considered useful to look at a few examples before moving onto the slightly more complicated parametric oscillator.

6. The Threshold for Amplification with Absorption

The most important threshold is when the output power is just equal to the input power and hence the power gain is unity.

From equation (58)

$$\exp((\beta_1 + \beta_2)l) = \frac{P^*P}{4\alpha\alpha^*} \sinh(\alpha l) \sinh(\alpha^* l) + \frac{P^*P}{2\alpha} \sinh(\alpha l) \cosh(\alpha^* l) + \frac{P^*P}{2\alpha^*} \cosh(\alpha^* l) \cosh(\alpha l) + \cosh(\alpha l) \cosh(\alpha^* l) \dots (5.64)$$

Now $P = i\Delta k - \beta_1 + \beta_2$... (5.65)

and $\alpha = \sqrt{\frac{P^2 + 4c_1c_1^*}{2}}$... (5.66)

The quantity c_1, c_1^* real:

$$c_1c_1^* = \frac{4\pi^2\omega_1^2\omega_2^2}{R_1R_2C^2\cos^2\alpha, \omega^2\alpha^2} \hat{E}_1 \cdot \hat{X}_1 \hat{E}_2^* \hat{X}_2^* \hat{C}_1 \hat{C}_2 A_1^* A_2 \dots (5.67)$$

and hence by solving the transcedental equation (64) with parameters $\beta_1, \beta_2, \Delta k$ specified it is possible to find the threshold power for unity power gain.

The equation is impossible to solve generally and so it must be solved by numerical, approximate techniques or for special cases.

Special case 1

$$\Delta k = 0, A = B$$

$$\text{then } \alpha = \alpha^* = \sqrt{C_1 C_2} \text{ and } \rho = 0$$

Equation (64) simplifies to

$$\exp((\beta_1 + \beta_2)l) = \cosh^2 \alpha l \quad \dots (5.68)$$

The approximation

$$\cosh^2 \alpha l = 1 + (\alpha l)^2 \quad \dots (5.69)$$

is now substituted to give

$$\alpha^2 = \frac{\exp(\beta_1 + \beta_2)l - 1}{l^2} \quad \dots (5.70)$$

Hence

$$I_{TH} = \frac{\exp(\beta_1 + \beta_2)l - 1}{l^2} R_1 R_2 \cos \alpha l \cos \alpha n, \dots (5.71)$$

This equation shows how the threshold pump power increases with the absorption coefficients. It is interesting to note that the signal and idler absorptions appear together.

7. The Parametric Oscillator

The parametric oscillator with no absorption at the signal and idler frequencies is, in fact, a special case of the oscillator when there is absorption at the signal and idler frequencies and in this chapter it will be treated as such. In the literature a distinction is made between the singly-resonant oscillator and the doubly-resonant oscillator that is oscillators which have one or two frequencies resonant. In the present work, the two are considered together by taking the reflectivities at the signal and idler frequencies to be parameters and later considering the special cases.

From equations (28) and (29)

$$A_1(z) = A_1^0 M(z) + A_1^{0*} N(z) \quad \dots (5.72)$$

$$A_2(z) = A_1^0 P(z) + A_2^{0*} Q(z) \quad \dots (5.73)$$

and M , N , P , Q are defined by equations (30) to (33).

A small signal, A_{in}^0 , is present at $z=0$, and this enters the crystal and at the end face of the crystal becomes $A_1(\ell)$ and $A_2(\ell)$ respectively. These are then reflected at $z = \ell$ and again at $z = 0$, when the amplitude is the same as at $z = 0$. Initially this process is :

$$A_1^0 = A_{in} + R_1^2 \exp(-\beta_1 \ell) \exp(i 2 k_1 \ell) A_1(\ell) \quad \dots (5.74)$$

$$A_2^{0*} = R_2^2 \exp(-\beta_2 \ell) \exp(i 2 k_2 \ell) A_2(\ell) \quad \dots (5.75)$$

The signal and idler frequencies are resonant hence:

$$\exp(i 2 k_1 \ell) = \exp(i 2 k_2 \ell) = 1 \quad \dots (5.76)$$

From (73) and (76)

$$A_2^{0*} = R_2^2 \exp(-\beta_2 \ell) (P(\ell) A_1^0 + Q(\ell) A_2^{0*}) \quad \dots (5.77)$$

$$A_2^{0*} (1 - R_2^2 \exp(-\beta_2 \ell) Q(\ell)) = R_2^2 \exp(-\beta_2 \ell) P(\ell) A_1^0 \quad \dots (5.78)$$

From (77) and (72)

$$A(\ell) = A_1^0 M(\ell) + \frac{N(\ell) R_1^2 \exp(-\beta_1 \ell) P(\ell) A_1^0}{1 - R_2^2 \exp(-\beta_2 \ell) Q(\ell)} \quad \dots (5.79)$$

Therefore $(1 - R_2^2 \exp(-\beta_2 \ell) Q(\ell)) A_1(\ell) = A_{in} \{ M(\ell) + R_1^2 \exp(-\beta_1 \ell) (N(\ell) P(\ell) - M(\ell) Q(\ell)) \}$

$+ R_2^2 \exp(-\beta_2 \ell) A_2(\ell) \{ M(\ell) + R_1^2 \exp(-\beta_1 \ell) (N(\ell) P(\ell) - M(\ell) Q(\ell)) \}$

$\dots (5.80)$

Now

$$P(\ell)N(\ell) = \exp(-(\beta_1 + \beta_2)\ell) \left\{ \frac{C_1 C_2}{\alpha^2} \sinh^2 \alpha \ell \right\} \quad (5.81)$$

$$M(\ell)Q(\ell) = \exp(-(\beta_1 + \beta_2)\ell) \left\{ \cosh^2 \alpha \ell - \frac{f^2}{4\alpha^2} \sinh^2 \alpha \ell \right\} \quad (5.82)$$

Hence

$$P(\ell)N(\ell) - M(\ell)Q(\ell) = - \exp(-(\beta_1 + \beta_2)\ell) \quad \dots \quad (5.83)$$

Thus from (80)

$$\frac{A_1(\ell)}{A_{in}} = \frac{M(\ell) - R_i^2 \exp(-(\beta_1 + 2\beta_2)\ell)}{1 - R_i^2 \exp(-\beta_1\ell)Q(\ell) - R_s^2 \exp(-\beta_1\ell)M(\ell) + R_s^2 R_i^2 \exp(-2\beta_1\ell)} \quad (5.84)$$

$$A_{in} = 1 - R_i^2 \exp(-\beta_1\ell)Q(\ell) - R_s^2 \exp(-\beta_1\ell)M(\ell) + R_s^2 R_i^2 \exp(-2\beta_1\ell)$$

The oscillator threshold is that pump power which is necessary for oscillations to be sustained with no injected signal.

It is determined by:

$$\frac{A_1(\ell)}{A_{in}} = \infty \quad \dots \quad (5.85)$$

i.e. when the denominator is zero.

$$1 - R_i^2 \exp(-\beta_1\ell)Q(\ell) - R_s^2 \exp(-\beta_1\ell)M(\ell) + R_s^2 R_i^2 \exp(-2(\beta_1 + \beta_2)\ell) = 0 \quad (5.86)$$

$$\begin{aligned} \text{i.e. } & 1 - R_i^2 \exp\left(-\frac{(\beta_1 + 3\beta_2)}{2}\ell\right) \exp\left(\frac{i\Delta k}{2}\ell\right) \left(\cosh \alpha \ell - \frac{f}{2\alpha} \sinh \alpha \ell \right) \\ & - R_s^2 \exp\left(-\frac{(3\beta_1 + \beta_2)}{2}\ell\right) \exp\left(\frac{i\Delta k}{2}\ell\right) \left(\cosh \alpha \ell + \frac{f}{2\alpha} \sinh \alpha \ell \right) \dots \quad (5.87) \\ & + R_s^2 R_i^2 \exp(-2(\beta_1 + \beta_2)\ell) \end{aligned}$$

Equation (87) is solved for α and the threshold determined. As can be seen this equation is not readily soluble and a solution under arbitrary conditions would call for numerical methods. However, it is possible to solve the equation under special conditions. In practice parametric oscillators are usually run

in a near-degenerate condition, that is with the signal and idler frequencies nearly equal, and in a nearly phase matched condition.

Because of the former it is a good approximation to set β_1 equal to β_2 if the signal and idler are of the same polarization and because of the latter it is useful to set Δk equal to zero.

The threshold condition then becomes greatly simplified.

From equation (87) can be obtained the singly- and doubly-resonant parametric oscillator cases. The singly-resonant case happens when one of the reflection coefficients, R_s or R_i , becomes zero.

8. Special Cases

Signal only resonant, i.e. $R_i = 0$

Equation (87) becomes

$$1 - R_s^2 \exp\left(-\left(\frac{3\beta_1 + \beta_2}{2}\right)l\right) \exp\left(-i\frac{\Delta k l}{2} \left(\cosh \alpha l + \frac{1}{2\alpha} \sinh \alpha l\right)\right) = 0. \quad (5.88)$$

and when $\beta_1 = \beta_2$ and $\Delta k = 0$

$$1 - R_s^2 \exp(-2\beta_1 l) (\cosh \alpha l) = 0 \quad \dots (5.89)$$

and

$$\alpha = \sqrt{C_1 C_2} \quad \dots (5.90)$$

Signal and idler resonant

$$\begin{aligned} & 1 - R_i^2 \exp\left(-\left(\frac{\beta_1 + 3\beta_2}{2}\right)l\right) \exp\left(i\frac{\Delta k l}{2} \left(\cosh \alpha l - \frac{1}{2\alpha} \sinh \alpha l\right)\right) \\ & - R_s^2 \exp\left(-\left(\frac{3\beta_1 + \beta_2}{2}\right)l\right) \exp\left(-i\frac{\Delta k l}{2} \left(\cosh \alpha l + \frac{1}{2\alpha} \sinh \alpha l\right)\right) \\ & + R_s^2 R_i^2 \exp(-2\beta_1 l) = 0 \quad \dots (5.91) \end{aligned}$$

When $\beta_1 = \beta_2$ and $\Delta k = 0$, (91) becomes

$$1 - R_i^2 \exp(-2\beta_i l) \left(\cosh \alpha l - \frac{1}{2\alpha} \sinh \alpha l \right) - R_s^2 \exp(-2\beta_i l) \left(\cosh \alpha l - \frac{1}{2\alpha} \sinh \alpha l \right) + R_s^2 R_i^2 \exp(-4\beta_i l) = 0 \quad (5.92)$$

The threshold is found by solving for α .

Determination of the threshold in special cases

No absorption, no mismatch case

$$\beta_1 = \beta_2 = 0 = \Delta k \quad \dots (5.93)$$

From (87)

$$1 - R_i^2 \cosh(\alpha l) - R_s^2 \cosh(\alpha l) + R_s^2 R_i^2 = 0 \quad \dots (5.94)$$

so

$$\cosh \alpha l = \frac{1 + R_s^2 R_i^2}{R_i^2 + R_s^2} \quad \dots (5.95)$$

$$(\alpha l)^2 = \left\{ \cosh^{-1} \left(\frac{1 + R_s^2 R_i^2}{R_i^2 + R_s^2} \right) \right\}^2 \quad \dots (5.96)$$

$$\therefore I_{TH} = \frac{R_s R_i c^4 \cos^2 \theta_i \cos^2 \theta_s n_3}{\ell^4 \omega^2 4\pi (e_1^2 e_2^2 e_3^2 e_4^2 e_5^2 e_6^2 e_7^2 e_8^2 e_9^2 e_1^2 e_2^2 e_3^2 e_4^2 e_5^2 e_6^2 e_7^2 e_8^2 e_9^2) \left[\frac{\cosh^{-1} \left(\frac{1 + R_s^2 R_i^2}{R_i^2 + R_s^2} \right)}{\left(R_s^2 + R_i^2 \right)^2} \right]^2} \quad (5.97)$$

An approximate solution of (95) can be obtained by expanding $\cosh(\alpha l)$ in a power series and ignoring all but the first two terms:

$$\frac{1 + R_s^2 R_i^2}{R_i^2 + R_s^2} = 1 + \frac{(\alpha l)^2}{2!}$$

then

$$\alpha^2 = \frac{2}{\ell^2} \left(\frac{1 + R_s^2 R_i^2 - R_i^2 - R_s^2}{R_i^2 + R_s^2} \right) \quad \dots (5.98)$$

Special case when $\beta_1 = \beta_2$ and $\Delta k = 0$

$$1 + R_s^2 R_i^2 \exp(-4\beta l) = \exp(-2\beta l) \cosh(\alpha l) (R_i^2 + R_s^2) \quad \dots (5.99)$$

$$\therefore \cosh \alpha l = \frac{\exp(2\beta l) + R_s^2 R_i^2 \exp(-2\beta l)}{R_i^2 + R_s^2} \quad \dots (5.100)$$

$$\cosh \alpha l = 1 + \frac{(\alpha l)^2}{2!} \dots \dots \dots (5.101)$$

$$\therefore (\alpha l)^2 = \frac{2 \exp(2\beta l) + R_i^2 R_s^2 \exp(-2\beta l) - R_i^2 - R_s^2}{R_i^2 + R_s^2} \quad \dots (5.102)$$

$$I_{TH} = \frac{k_1 R_i c^4 \cos^2 \alpha \cos^2 \alpha \cdot n_1}{\cancel{\omega_1 \omega_2 \omega_3} \cancel{\omega_1^2 \omega_2^2 \omega_3^2} \cancel{6\pi} (\vec{e}_1 \cdot \vec{e}_2; \vec{e}_3; \vec{e}_1 \cdot \vec{e}_2; \vec{e}_3) \alpha^2} \quad \dots (5.103)$$

APPENDIX I DERIVATION OF A VECTOR RELATIONSHIP

In this appendix is derived an expression for $\hat{F} \cdot \nabla \times \nabla \times \underline{F}$ where \underline{F} represents a vector wave propagating in the $+z$ direction and $\hat{F} = \frac{\underline{F}}{|\underline{F}|}$

For a wave which is infinite in the x , y plane the operations $\partial/\partial x$ and $\partial/\partial y$ are equivalent to multiplication by zero then

$$\nabla \times \underline{F} = \begin{vmatrix} \hat{i} & \hat{j} & \hat{k} \\ 0 & 0 & \frac{\partial}{\partial z} \\ F_x & F_y & F_z \end{vmatrix} = \hat{i} \left(-\frac{\partial F_y}{\partial z} \right) + \hat{j} \left(\frac{\partial F_x}{\partial z} \right)$$

thus

$$\nabla \times \nabla \times \underline{F} = \begin{vmatrix} \hat{i} & \hat{j} & \hat{k} \\ 0 & 0 & \frac{\partial}{\partial z} \\ -\frac{\partial F_y}{\partial z} & \frac{\partial F_x}{\partial z} & 0 \end{vmatrix} = -\frac{\partial^2}{\partial z^2} (\hat{i} F_x + \hat{j} F_y)$$

hence

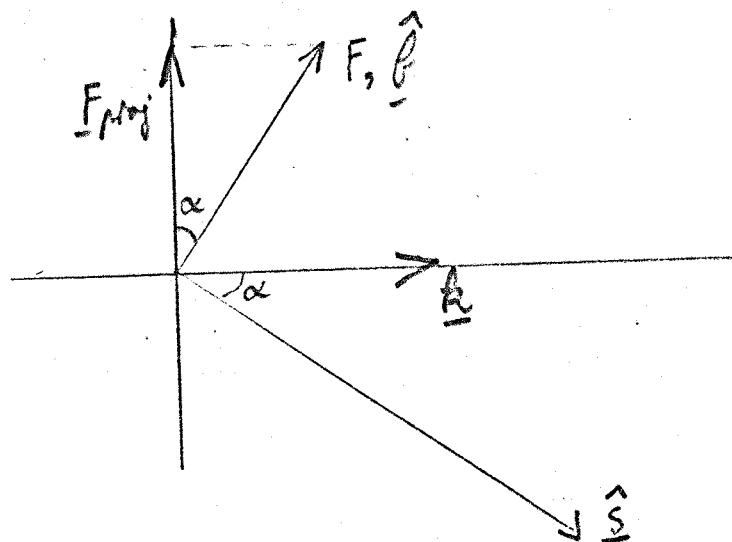
$\hat{F} \cdot \nabla \times \nabla \times \underline{F} = -\hat{F} \cdot \frac{\partial^2}{\partial z^2} (\hat{i} F_x + \hat{j} F_y) = -\frac{\partial}{\partial z^2} (\hat{F} \cdot (\hat{i} F_x + \hat{j} F_y))$
 from diagram 1 of the vector $\hat{i} F_x + \hat{j} F_y = F_{\text{proj}}$ is the vector in the $z=0$ plane which is the projection of \underline{F} in this plane; it is equal in magnitude to $|\underline{F}| \cos \alpha$. ($|\underline{F}|$ is a complex number)

hence $\hat{F} \cdot (\hat{i} F_x + \hat{j} F_y) = |\underline{F}| \cos^2 \alpha$

hence $\hat{F} \cdot \nabla \times \nabla \times \underline{F} = -\cos^2 \alpha \frac{\partial^2 |\underline{F}|^2}{\partial z^2}$

In our case the angle α is the angle between the wave normal direction and the Poynting vector.

Diagram 1



\hat{s} is the direction perpendicular to \underline{F} in the same plane as \underline{k} and \underline{E} . The direction \underline{F} corresponds to that of E , F_{Mag} to that of \underline{D} , \underline{k} to the wave normal direction and \underline{s} to the direction of the Poynting vector.

APPENDIX II TO DERIVE AN EXPRESSION FOR E D

This derivative applies to crystals which possess anisotropy of the dielectric tensor and optical activity, but which are non-absorbing.

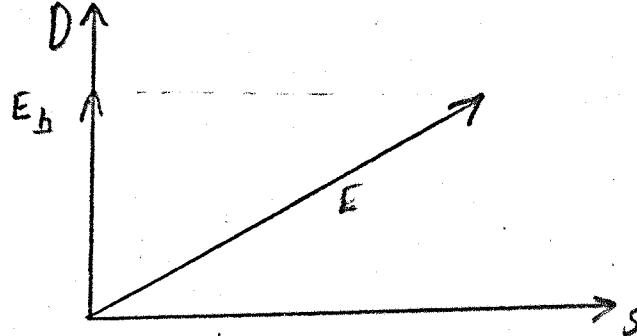
We start with Maxwell's equations and consider infinite plane waves. The field vectors \underline{E} , \underline{D} , \underline{H} and \underline{B} are proportional to $\exp[i\omega\left(\frac{n}{c}(t, s) - t\right)]$ and hence the operations $\partial/\partial t$ and $\partial/\partial x$ are equivalent to multiplication by $-i\omega$ and $i\omega n s_x/c$ respectively.

Hence $n \underline{s} \times \underline{H} = -\underline{D}$ and $n \underline{s} \wedge \underline{E} = \underline{H}$ become Maxwell's curl equations. ... (1)

Eliminating \underline{H} between equations (1) and then using a well known vector identity gives:

$$\underline{D} = -n^2 \underline{s} \times (\underline{s} \times \underline{E}) = n^2 [\underline{E} - \underline{s} (\underline{s} \cdot \underline{E})] = n^2 \underline{E}_h \quad \dots (2)$$

Figure 1.



Here \underline{E}_h is the vector component of \underline{E} in the direction of \underline{D} . See fig.1.

... (3)

Thus from (2)

$$\underline{D} = \frac{n^2 (\underline{E} \cdot \underline{D}) \underline{D}}{|\underline{D}| |\underline{D}|} \quad \dots (4)$$

Now

$$\underline{E} \cdot \underline{D} = |\underline{E}| |\underline{D}| \cos \alpha \quad \dots (5)$$

so

$$|\underline{D}| |\underline{D}| = n^2 |\underline{E}| |\underline{D}| \cos \alpha \quad \dots (6)$$

Multiply both sides of this expression by $\frac{|\underline{E}| \cos \alpha}{|\underline{D}|}$

which gives:

$$|\underline{E}| |\underline{D}| \cos \alpha = n^2 |\underline{E}| |\underline{E}| \cos^2 \alpha \quad \dots (7)$$

From (5)

$$(\underline{E} \cdot \underline{D}) = n^2 \cos^2 \alpha |\underline{E}|^2$$

and

$$\hat{\underline{e}} \cdot \underline{D} = n^2 \cos^2 \alpha |\underline{E}|$$

where

$\hat{\underline{e}}$ is the unit vector of \underline{E}

APPENDIX III DERIVATION OF AN EXPRESSION FOR $E \cdot (D + \frac{i4\pi}{\omega} \sigma \cdot E)$

This identity is for crystals which are both anisotropic and which are anisotropic in their absorption coefficient.

We start from Maxwell's equations and consider infinite plane waves. The field vectors \underline{E} , \underline{D} , \underline{H} and \underline{B} are proportional to $\exp[i\omega(\frac{n'}{c}(t,s)-t)]$ and hence the operations $\partial/\partial t$ and $\partial/\partial x$ are equivalent to multiplication by $-i\omega$ and $i\omega n S_x/c$ respectively. n' is not the complex refractive index $n' = n - i\alpha$, where n is the part which govern the phase velocity and α is the absorption coefficient at that particular frequency.

Hence from Maxwell's curl equations :

$$n \underline{S} \times \underline{H} = -(\underline{D} + i\frac{4\pi}{\omega} \underline{\sigma} \cdot \underline{E}) \quad \dots (1)$$

and $n \underline{S} \times \underline{E} = \underline{H} \quad \dots (2)$

Eliminating \underline{H} between (1) and (2) gives:

$$\begin{aligned} \underline{D} + i\frac{4\pi}{\omega} \underline{\sigma} \cdot \underline{E} &= -n^2 \underline{S} \times (\underline{S} \times \underline{E}) && \text{from a well known} \\ &= n^2 [\underline{E} - \underline{S}(\underline{S} \cdot \underline{E})] && \text{vector identity} \\ &= n^2 \underline{E}_B \end{aligned} \quad \dots (3)$$

see fig. 2.

Here \underline{E}_B is the vector component of \underline{E} in the direction of $\underline{D} + i\frac{4\pi}{\omega} \underline{\sigma} \cdot \underline{E}$

$$\underline{E}_B = \underline{E} \cdot \left(\underline{D} + \frac{i4\pi}{\omega} \underline{\sigma} \cdot \underline{E} \right) \left(\underline{D} + \frac{i4\pi}{\omega} \underline{\sigma} \cdot \underline{E} \right) \quad \dots (4)$$

$$\left| \underline{D} + \frac{i4\pi}{\omega} \underline{\sigma} \cdot \underline{E} \right| \left| \underline{D} + \frac{i4\pi}{\omega} \underline{\sigma} \cdot \underline{E} \right|$$

Hence from (3)

$$\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E} = n^2 \underline{E} \cdot \frac{(\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E})(\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E})}{(\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E})^2} \dots (5)$$

Thus

$$|\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}|^2 = n^2 \underline{E} \cdot (\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}) \dots (6)$$

Now

$$\underline{E} \cdot (\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}) = |\underline{E}| |\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}| \cos \alpha \dots (7)$$

So from (5)

$$|\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}|^2 = n^2 |\underline{E}| |\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}| \cos \alpha$$

Multiply both sides of this expression by $\frac{|\underline{E}| \cos \alpha}{(\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E})}$
then

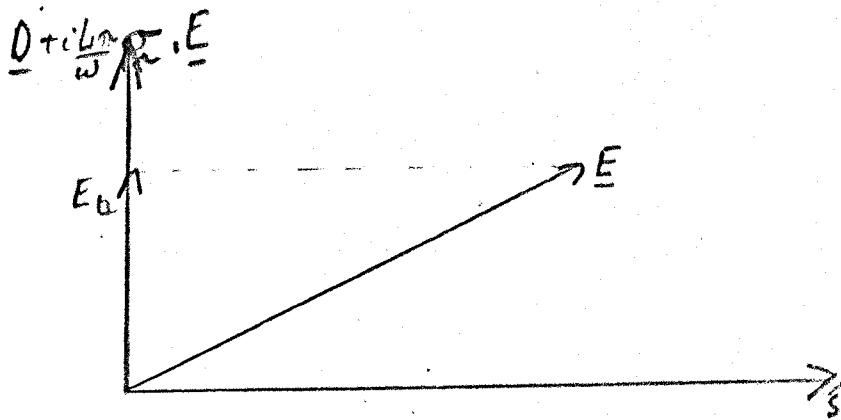
$$|\underline{E}| \cdot |\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}| \cos \alpha = n^2 |\underline{E}|^2 \cos^2 \alpha$$

Again using (7)

$$\underline{E} \cdot (\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}) = n^2 \cos^2 \alpha |\underline{E}|^2$$

hence

$$\hat{\underline{E}} \cdot (\underline{D} + i \frac{4\pi}{\omega} \sigma \cdot \underline{E}) = n^2 \cos^2 |\underline{E}|$$



REFERENCES

Szivessy 1928 : G. Szivessy, Kristalloptik, Handbuch der Physik, Vol.20, Berlin Springer 1928

Ram. and Ram. 1961 : G. N. Ramachandran and S. Ramaseshan, Crystal Optics, Handbuch der Physik, Band XXV/1, Berlin Springer 1961

Born and Wolf 1965 : 'Principles of Optics', Pergamon Press 1965

Franken and Ward 1963 : P. A. Franken and J. F. Ward, 'Optical Harmonics and Nonlinear Phenomena', Rev. Mod. Phys. 35, 1, 23

ABDP 1962 : J. A. Armstrong, N. Bloembergen, J. Ducuing and P.S. Pershan, 'Interactions between Light Waves in a Nonlinear Dielectric', Phys. Rev., 127, 6, 1918

Rabin and Bey 1967 : H. Rabin and P. B. Bey, 'Phase Matching in Harmonic Generation Employing Optical Rotatory Dispersion', Phys. Rev., 156, 1967, 1010

Rabin and Bey 1967 : H. Rabin and P. B. Bey, 'Coupled Wave Solution of Harmonic Generation in an Optically Active Medium', Phys. Rev., 162, 1967, 794

Bloem 1964 : N. Bloembergen, 'Nonlinear Optics', Benjamin 1964

Yariv 1967 : A. Yariv, 'Quantum Electronics', Wiley 1967

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