Excerpts from “Handbook of nonlinear optics”
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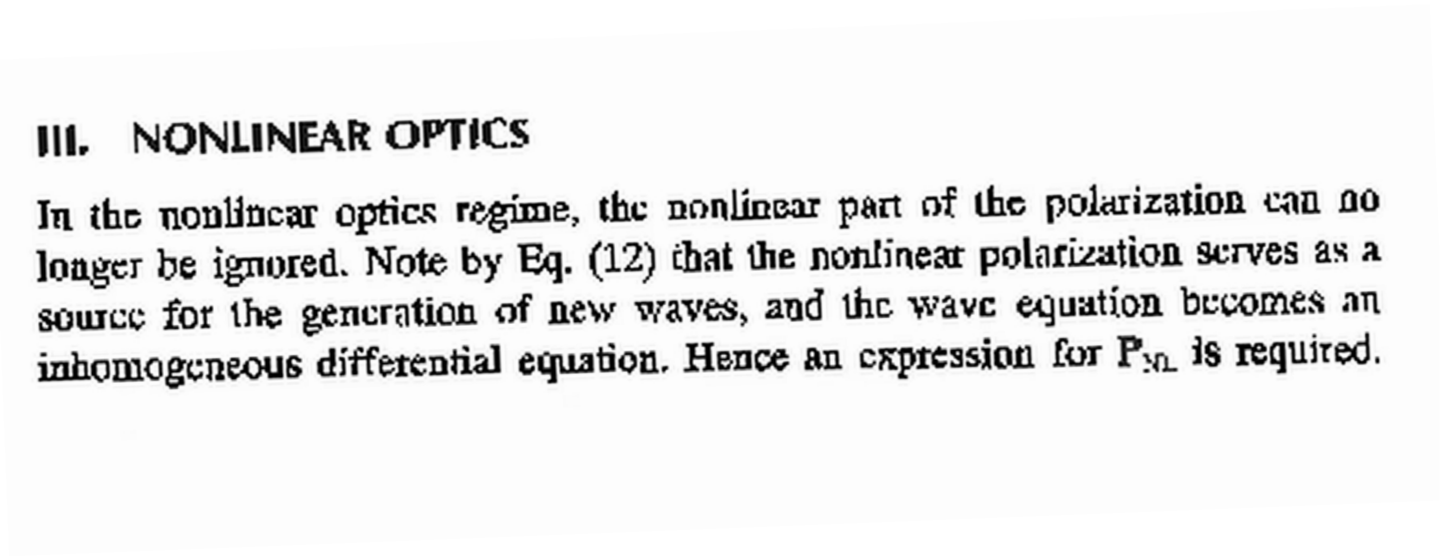

III. NONLINEAR OPTICS

In the nonlinear optics regime, the nonlinear part of the polarization can no longer be ignored. Note by Eq. (12) that the nonlinear polarization serves as a source for the generation of new waves, and the wave equation becomes an inhomogeneous differential equation. Hence an expression for $\mathbf{P}_{NL}$ is required.

For most of the applications of nonlinear optics, this quantity can be expressed as a power series expansion in the applied fields.

A. Nonlinear Susceptibilities

It is assumed that the nonlinear polarization can be written as [4–6]

$$\mathbf{P}_{NL}^{(t)} = \mathbf{P}^{(t)(2)} + \mathbf{P}^{(t)(3)} + \dots \tag{39}$$

where

$$\mathbf{P}^{(t)(2)} = \varepsilon_0 \boldsymbol{\chi}^{(2)} : \mathbf{E}^{(t)}\mathbf{E}^{(t)} \tag{40}$$

$$\mathbf{P}^{(t)(3)} = \varepsilon_0 \boldsymbol{\chi}^{(3)} \vdots \mathbf{E}^{(t)}\mathbf{E}^{(t)}\mathbf{E}^{(t)} \tag{41}$$

$$\vdots$$

etc.

These expressions are given in SI units. For cgs units, $\varepsilon_0 \rightarrow 1$. It is important to note that the field in the equations above is the *total* applied field, which can be a superposition of many fields of different frequencies. $\chi^{(n)}$ is the nth order dielectric response and is a tensor of rank $n + 1$.

When the total field is expanded in terms of its Fourier components (e.g., its various laser frequencies), then the nonlinear polarization will consist of several terms oscillating at various combination frequencies. For example, if the total field consists of two waves oscillating at frequencies ω_1 and ω_2, the second order nonlinear polarization will have components oscillating at $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$, $\omega_1 - \omega_2$, and dc terms at zero frequency. Similarly, with three fields oscillating at frequencies ω_1, ω_2, and ω_3, the third order polarization will oscillate at $3\omega_1$, $3\omega_2$, $3\omega_3$, $\omega_1 + \omega_2 + \omega_3$, $\omega_1 + \omega_2 - \omega_3$, etc.

It is common to write the Fourier components of the nonlinear polarization in the following way. Consider a second order polarization oscillating at ω_3 due to the presence of fields oscillating at frequencies ω_1 and ω_2, with $\omega_3 = \omega_1 + \omega_2$. Then the ith cartesian component of the complex polarization amplitude is expressed as

$$P_i^{(2)}(\omega_3) = \varepsilon_0 D^{(2)} \sum_{jk} \chi_{ijk}^{(2)}(-\omega_3; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2) \tag{42}$$

where

$$D^{(2)} = \begin{cases} 1 & \text{for indistinguishable fields} \\ 2 & \text{for distinguishable fields} \end{cases} \tag{43}$$

and $\chi_{ijk}^{(2)}(-\omega_3;\omega_1,\omega_2)$ is the second order (complex) Fourier-transformed dielectric response, or the second order susceptibility. The form of Eq. (42) allows for the possibility that the frequencies ω_1 and ω_2 are equal, or equal in magnitude and opposite in sign. In this case, there may actually be only one field present, and the degeneracy factor $D^{(2)}$ takes this into account. It should be noted, however, that the determination of the degeneracy factor is whether the fields are physically distinguishable or not. Two fields of the same frequency will be physically distinguishable if they travel in different directions, for example. Also, the negative frequency part of the real field is considered to be distinguishable from the positive frequency part, i.e., they have different frequencies. For negative frequencies, it is important to note that $E(-\omega) = E^*(\omega)$, since the rapidly varying field is a real mathematical quantity. Thus, for example, if $\omega_1 = \omega$ and $\omega_2 = -\omega$, then the second order polarization would be written as

$$P_i^{(2)}(0) = 2\epsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(0;\omega,-\omega)E_j(\omega)E_k^*(\omega) \tag{44}$$

This polarization drives the phenomenon known as optical rectification, wherein an intense optical wave creates a dc polarization in a nonlinear medium. It is important to remember that in these equations, the field amplitude still contains the rapidly varying spatial part, i.e., $E(\mathbf{r}) = A(\mathbf{r})\exp(i\mathbf{k}\cdot\mathbf{r})$.

This notation is easily extended to higher orders. When three frequencies ω_1, ω_2, ω_3 are present, the third order polarization at $\omega_4 = \omega_1 + \omega_2 + \omega_3$ is given by

$$P_i^{(3)}(\omega_4) = \epsilon_0 D^{(3)} \sum_{jkl} \chi_{ijkl}^{(3)}(-\omega_4;\omega_1,\omega_2,\omega_3)E_j(\omega_1)E_k(\omega_2)E_l(\omega_3) \tag{45}$$

where the degeneracy factor in this case becomes

$$D^{(3)} = \begin{cases} 1 & \text{all fields indistinguishable} \\ 3 & \text{two fields indistinguishable} \\ 6 & \text{all fields distinguishable} \end{cases} \tag{46}$$

This form of the third order polarization allows for various combination frequencies even when only two fields are present, such as $\omega_1 + 2\omega_2$, or $2\omega_1 - \omega_2$, etc. The degeneracy factor is just due to the number of different ways in which the products of the field Fourier components appear in the expansion of the total field to some power. For example, there is only one way that the product for the frequency $3\omega_1$ appears: $E(\omega_1)E(\omega_1)E(\omega_1)$. However, there are three different ways that the product for the frequency $2\omega_1 - \omega_2$ appears: $E(\omega_1)E(\omega_1)E^*(\omega_2)$, $E(\omega_1)E^*(\omega_2)E(\omega_1)$, and $E^*(\omega_2)E(\omega_1)E(\omega_1)$. The degeneracy factor is thus related to the coefficients of Pascal's triangle from algebra. (The use of a degeneracy factor in these equations also relies on an intrinsic symmetry of the susceptibility tensor, which is discussed below.)

The equations above are written in SI units. To obtain the form in cgs units let ϵ_0 go to 1. Also, it is important to note that the nth order susceptibility is frequency dependent and complex, in general. The reason for writing the frequency dependence as shown in the equations above is for the purpose of expressing symmetry relations of the susceptibility tensor. This is further described below.

B. Symmetry Relations of the Nonlinear Susceptibility

The first symmetry apparent from the form of Eqs. (42) and (45) is due to the lack of difference physically in which order the product of the field amplitudes is given. Thus an interchange in the order of the product $E_j(\omega_1)E_k(\omega_2)$ [i.e., $E_j(\omega_1)E_k(\omega_2) \leftrightarrow E_k(\omega_2)E_j(\omega_1)$] will not affect the value or sign of the ith component of the nonlinear polarization. The nonlinear susceptibility should reflect this symmetry. But note that in the above interchange, both frequencies and subscripts for the cartesian coordinates are interchanged simultaneously. This is important since, for example, exchanging the product $E_x(\omega_1)E_y(\omega_2)$ with the product $E_x(\omega_2)E_y(\omega_1)$ *could change the nonlinear polarization*, especially, for example, if the two fields are orthogonally polarized. Thus the symmetry property is expressed as (for third order susceptibilities)

$$\begin{aligned}\chi^{(3)}_{ijkl}(-\omega_4; \omega_1, \omega_2, \omega_3) &= \chi^{(3)}_{ikjl}(-\omega_4; \omega_2, \omega_1, \omega_3) \\ &= \chi^{(3)}_{iljk}(-\omega_4; \omega_3, \omega_2, \omega_1) = \text{etc.}\end{aligned} \tag{47}$$

In other words, if any of the subscripts $\{jkl\}$ are permuted, then the susceptibility will remain unchanged as long as the corresponding set of subscripts $\{123\}$ are also permuted. This holds even if any of the frequencies are negative. Note that this does not hold for the subscript pair $(i, 4)$. The same relation holds for second order and can be generalized to any order. This is called intrinsic permutation symmetry and is the underlying reason why the nonlinear polarization can be written compactly in terms of a degeneracy factor as in Eqs. (42) and (45).

At this point it is important to note another notation that is used in second order nonlinear optics. Often the susceptibility is represented as the so-called d-coefficient, where $\mathbf{d}$ is a tensor given by

$$d_{ijk} = \tfrac{1}{2}\chi^{(2)}_{ijk} \tag{48}$$

Furthermore, the intrinsic permutation symmetry is used to contract the last two subscripts and write $d_{ijk} \rightarrow d_{il}$. The subscripts are then written as numbers instead of letters using the scheme

$$\begin{array}{llll} i\!: & x\ 1 & jk\!: & xx \quad l\!:\ 1 \\ & y\ 2 & & yy \qquad\ \ 2 \\ & z\ 3 & & zz \qquad\ \ 3 \\ & & & yz = zy \quad 4 \\ & & & xz = zx \quad 5 \\ & & & xy = yx \quad 6 \end{array} \tag{49}$$

For example, $d_{xyz} = d_{xzy} = d_{14}$ and $d_{zxx} = d_{31}$, etc. The utility of this notation is that d-coefficients can be expressed as elements of a 3×6 matrix rather than a $3 \times 3 \times 3$ tensor. To use these coefficients in the nonlinear polarization of Eq. (42), just make the substitution $\chi_{ijk} \rightarrow 2d_{il}$.

Another form of the permutation symmetry can be shown when the nonlinear susceptibility is calculated quantum mechanically. This is usually done using the density matrix method, and expressions can be found in textbooks [4,5]. It can be shown generally, for example in third order, that

$$\chi^{(3)}_{ijkl}(-\omega_4; \omega_1, \omega_2, \omega_3) = \chi^{(3)*}_{jikl}(-\omega_1; \omega_4, -\omega_2, -\omega_3) = \text{etc.} \tag{50}$$

First, it is noted that when the complex conjugate of the susceptibility is taken, it just changes the sign of all of the frequencies. This is a consequence of the fact that the rapidly varying nonlinear polarization is a real mathematical quantity. Then, for *any* permutation of the cartesian subscripts, the new susceptibility thus obtained is equal to the original susceptibility if the corresponding frequency subscripts are also permuted, if all of the frequencies are changed by multiplying by -1, and if the complex conjugate of susceptibility is taken. This important result states that the susceptibilities for different physical processes are simply related. For example, Eq. (50) relates the susceptibilities for the third order processes of sum-frequency generation ($\omega_4 = \omega_1 + \omega_2 + \omega_3$) and difference-frequency generation ($\omega_1 = \omega_4 - \omega_2 - \omega_3$). The relations in Eq. (50) hold for any permutation and are generalized to all orders. Thus, for example, in second order,

$$\chi^{(2)}_{ijk}(-\omega_3; \omega_1, \omega_2) = \chi^{(2)*}_{kji}(-\omega_2; -\omega_1, \omega_3) = \text{etc.} \tag{51}$$

The above expressions hold generally when the susceptibilities are complex. This is especially important when any single frequency or any combination frequency is near a natural resonance frequency of the material. However, it is often the case in many applications that all frequencies and combination frequencies are far from any material resonance. Then the susceptibilities can be treated as real quantities, and any susceptibility is thus equal to its complex conjugate. In this case, Eq. (50) may be written as

$$\begin{aligned}\chi^{(3)}_{ijkl}(-\omega_4; \omega_1, \omega_2, \omega_3) &= \chi^{(3)}_{jikl}(-\omega_1; \omega_4, -\omega_2, -\omega_3) \\ &= \chi^{(3)}_{kjil}(-\omega_2; -\omega_1, \omega_4, -\omega_3) = \text{etc.}\end{aligned} \tag{52}$$

Thus, under the condition that the susceptibilities are real (all frequencies far from any resonance), the susceptibilities are unchanged for the simultaneous permutation of subscripts from the cartesian set $\{ijkl\}$ and the corresponding subscripts from the frequency set $\{4123\}$, with the stipulation that the frequencies carry the proper sign. (*Note:* The first frequency in the argument, that is, the *generated* frequency, carries a negative sign. The signs on the other frequencies must be such that the algebraic sum of all frequencies is zero. For example, $-\omega_4 + \omega_3 + \omega_2 + \omega_1 = 0$ implies that $-\omega_2 - \omega_1 + \omega_4 - \omega_3 = 0$.) This is called full permutation symmetry. This symmetry generalizes to all orders.

Another symmetry follows when the susceptibilities exhibit negligible dispersion over the entire frequency range of interest. Thus, in addition to all frequencies being far from any material resonance, this symmetry requires that there be no resonance between any of the frequencies. This would *not* be the case if, for example, $\omega_2 > \omega_1$ and for some frequency between ω_1 and ω_2 an absorption line exists in the material. When dispersion can be ignored, the frequencies can be freely permuted without permuting the corresponding cartesian subscripts, and vice versa, and the susceptibility will remain unchanged. This is known as Kleinman symmetry.

Nonlinear susceptibilities also reflect the structural symmetry of the material. This is important since in many cases this greatly reduces the number of nonzero, independent tensor components needed to describe the medium. One immediate consequence of this is that for all materials that possess a center of inversion symmetry (e.g., isotropic liquids and crystals of symmetry class 432), all elements of all even-order susceptibility tensors are identically equal to zero. Hence no even-order nonlinear processes are possible in these types of materials. (This is strictly true only when the susceptibility is derived using the electric dipole approximation in the perturbation Hamiltonian. For example, in an atomic vapor a second order process may occur when a transition matrix element between two equal parity states, which is forbidden in the electric dipole approximation, is nonzero due to an electric quadrupole allowed transition. However, such a transition is generally very weak.)

Other simplifications of the susceptibility tensors can be derived based on specific symmetry properties of the material, such as rotation axis and mirror plane symmetries. The forms of the *d*-matrix for crystals of several different symmetry classes are given in Table 1. Note that in many cases the elements are zero or equal to ±1 times other elements. The form of the matrix when Kleinman symmetry is valid is also given.

The specific form of third order susceptibilities may also be given for each symmetry class. These can be found in some textbooks [4,5]. In this book, third order effects are considered primarily in centrosymmetric media. The nonzero, independent elements of the third order susceptibility for materials in this class are shown in Table 2.

C. Coupled-Wave Propagation

The optical waves are coupled through the nonlinear polarization, and the nonlinear polarization acts as a source term in the wave equation for each monochromatic, or quasi-monochromatic, wave. As stated earlier, because of the

Table 1 Form of the d-Matrix for Different Symmetry Classes

Symmetry class	General conditions	Kleinman symmetry
	Biaxial crystals	
1	$\begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix}$	$\begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix}$
2	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & d_{16} \\ d_{21} & d_{22} & d_{23} & 0 & d_{25} & 0 \\ 0 & 0 & 0 & d_{34} & 0 & d_{36} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & d_{16} \\ d_{16} & d_{22} & d_{23} & 0 & d_{14} & 0 \\ 0 & 0 & 0 & d_{23} & 0 & d_{14} \end{pmatrix}$
m	$\begin{pmatrix} d_{11} & d_{12} & d_{13} & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & d_{26} \\ d_{31} & d_{32} & d_{33} & 0 & d_{35} & 0 \end{pmatrix}$	$\begin{pmatrix} d_{11} & d_{12} & d_{13} & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & d_{12} \\ d_{15} & d_{24} & d_{33} & 0 & d_{13} & 0 \end{pmatrix}$
222	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix}$
$mm2$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{15} & d_{24} & d_{33} & 0 & 0 & 0 \end{pmatrix}$
	Uniaxial crystals	
3	$\begin{pmatrix} d_{11} & -d_{11} & 0 & d_{14} & d_{15} & -d_{22} \\ d_{22} & d_{22} & 0 & d_{15} & -d_{14} & -d_{11} \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} d_{11} & -d_{11} & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & -d_{11} \\ d_{15} & d_{15} & d_{33} & 0 & 0 & 0 \end{pmatrix}$
$3m$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & 0 \\ d_{15} & d_{15} & d_{33} & 0 & 0 & 0 \end{pmatrix}$
$\overline{6}$	$\begin{pmatrix} d_{11} & -d_{11} & 0 & 0 & 0 & -d_{22} \\ -d_{22} & d_{22} & 0 & 0 & 0 & -d_{11} \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} d_{11} & -d_{11} & 0 & 0 & 0 & -d_{22} \\ -d_{22} & d_{22} & 0 & 0 & 0 & -d_{11} \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$

Table 1 Continued

Symmetry class	General conditions	Kleinman symmetry
	Uniaxial crystals	
$\bar{6}m2$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & -d_{22} \\ d_{22} & d_{22} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & -d_{22} \\ d_{22} & d_{22} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$
6,4	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & -d_{14} & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & 0 & 0 \\ d_{15} & d_{15} & d_{33} & 0 & 0 & 0 \end{pmatrix}$
$6mm,4mm$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & 0 & 0 \\ d_{15} & d_{15} & d_{33} & 0 & 0 & 0 \end{pmatrix}$
622,422	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & -d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$
$\bar{4}$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & d_{14} & 0 \\ d_{31} & -d_{31} & 0 & 0 & 0 & d_{36} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & d_{15} & 0 \\ 0 & 0 & 0 & -d_{15} & d_{14} & 0 \\ d_{15} & -d_{15} & 0 & 0 & 0 & d_{14} \end{pmatrix}$
32	$\begin{pmatrix} d_{11} & -d_{11} & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & -d_{14} & -d_{11} \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} d_{11} & -d_{11} & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & -d_{14} & -d_{11} \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$
$\bar{4}2m$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix}$
	Isotropic crystals	
$\bar{4}3m,23$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix}$
432	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$

Table 2 Form of the Third Order Susceptibility for Centrosymmetric Media

$$\chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} = \chi^{(3)}_{xxzz} = \chi^{(3)}_{zzxx} = \chi^{(3)}_{yyzz} = \chi^{(3)}_{zzyy}$$

$$\chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx} = \chi^{(3)}_{xzxz} = \chi^{(3)}_{zxzx} = \chi^{(3)}_{yzyz} = \chi^{(3)}_{zyzy}$$

$$\chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy} = \chi^{(3)}_{xzzx} = \chi^{(3)}_{zxxz} = \chi^{(3)}_{yzzy} = \chi^{(3)}_{zyyz}$$

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz}$$

All other components are zero. There are only three nonzero independent components, and

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx}$$

linearity of the wave equation, each frequency component of the field satisfies the wave equation independently, with the source term being the Fourier component of the nonlinear polarization corresponding to the frequency of that optical field.

In this section, the form of the coupled-wave equations is considered when the waves propagate in an isotropic medium or along one of the principal axes in an anisotropic medium. When the birefringence of a uniaxial medium is small, which is quite often true in applications, the forms of these equations will be the same to a good approximation for propagation along any axis in a uniaxial medium.

Consider the interaction of $m + 1$ waves through an mth order nonlinear polarization. The frequency of the $(m + 1)$th wave is given by

$$\omega_{m+1} = \sum_{\mu=1}^{m} \omega_\mu \tag{53}$$

where the set $\{\omega_\mu\}$ may contain both positive and negative frequencies. For any wave amplitude, $A(-\omega) = A^*(\omega)$, and for any wave vector, $k(-\omega) = -k(\omega)$. The wave amplitudes are assumed to vary primarily along the z-axis.

For practically every problem in nonlinear optics, the slowly varying amplitude approximation may be used [4–6]. This assumes that the magnitude and phase of the wave amplitude vary slowly in space and time over an optical wavelength and period, respectively. For any wave amplitude this implies that

$$\left|\frac{\partial^2 A}{\partial z^2}\right| \ll \left|k \frac{\partial A}{\partial z}\right| \tag{54}$$

$$\left|\frac{\partial^2 A}{\partial t^2}\right| \ll \left|\omega \frac{\partial A}{\partial t}\right| \tag{55}$$

and for the complex amplitude of the Fourier component of the nonlinear polarization,

$$\left|\frac{\partial^2 \mathbf{P}^{(m)}}{\partial t^2}\right| \ll \left|\omega \frac{\partial \mathbf{P}^{(m)}}{\partial t}\right| \ll \left|\omega^2 \mathbf{P}^{(m)}\right| \tag{56}$$

In all of the wave equations given below, the following definitions apply:

$$\chi_{\text{eff}}^{(m)} = \hat{e}_{m+1} \cdot \boldsymbol{\chi}^{(m)}(-\omega_{m+1}; \omega_1, \omega_2, \ldots, \omega_m) \vdots \hat{e}_1 \hat{e}_2 \cdots \hat{e}_m \tag{57}$$

where $\hat{e}_\mu$ is a unit vector pointing in the direction in space of the polarization of the μth field,

$$\Delta k = \left(\sum_{\mu=1}^{m} \mathbf{k}_\mu - \mathbf{k}_{m+1}\right) \cdot \hat{z} \tag{58}$$

$$K' = \begin{cases} 1 & \text{(SI)} \\ 4\pi & \text{(cgs)} \end{cases} \tag{59}$$

and

$$n_{m+1} = n(\omega_{m+1}) \tag{60}$$

$$\alpha_{m+1} = \alpha(\omega_{m+1}) \tag{61}$$

are the frequency dependent index of refraction and absorption coefficient, respectively.

The following equations are the form of the wave equation for A_{m+1} encountered under various situations. The terms quasi-cw, short pulse, and ultra-short pulse have the following meanings. Quasi-cw may refer to a true cw wave or to a pulse with a full width at half-maximum (t_{FWHM}) such that the physical length of the medium is small compared to the distance ct_{FWHM}/n. A short pulse will have a width such that the physical length of the medium is comparable to or larger than ct_{FWHM}/n. An ultra-short pulse is only quasi-monochromatic and hence is composed of several frequencies clustered about some center frequency. When the frequency spread of the pulse is such that dispersion in the refractive index cannot be ignored, the concept of a unique phase velocity is meaningless. The pulse is then treated as a superposition of monochromatic waves clustered about the center frequency, which move as a group with group velocity $v_g = (dk/d\omega)^{-1}$. This term is encountered when the physical length of the medium is comparable to or larger than the distance $v_g t_{\text{FWHM}}$.

The wave equation is thus given by

1. *Infinite plane waves, no absorption, quasi-cw, propagation primarily along* $+z$:

$$\frac{dA_{m+1}}{dz} = i\,\frac{K'\omega_{m+1}}{2n_{m+1}c}\,D^{(m)}\chi_{\text{eff}}^{(m)} \prod_{\mu=1}^{m} A_\mu \exp(i\Delta kz) \tag{62}$$

Table 1 Units of Important Physical Parameters Used in Frequency Conversion Formulas

Parameter	SI units	cgs units
Second order polarization	$[P^{(2)}] = \mathrm{C/m^2}$	$[P^{(2)}] = \mathrm{sC/cm^2} = \mathrm{dyn^{1/2}/cm}$ $= (\mathrm{erg/cm^3})^{1/2}$
Electric field	$[E] = \mathrm{V/m}$	$[E] = \mathrm{sV/cm} = (\mathrm{erg/cm^3})^{1/2}$
Second order susceptibility	$[d] = \mathrm{m/V}$	$[d] = \mathrm{cm^2/sC} = \mathrm{cm/dyn^{1/2}}$ $= (\mathrm{cm^3/erg})^{1/2} = \mathrm{esu}$
Vacuum permittivity	$[\varepsilon_0] = \mathrm{F/m} = \mathrm{C/V \cdot m}$	—
Distance, length	$[z] = [r] = [L] = \mathrm{m}$	$[z] = [r] = [L] = \mathrm{cm}$
Speed of light	$[c] = \mathrm{m/s}$	$[c] = \mathrm{cm/s}$
Wavelength	$[\lambda] = \mathrm{m}$	$[\lambda] = \mathrm{cm}$
Wave vector	$[k] = \mathrm{m^{-1}}$	$[k] = \mathrm{cm^{-1}}$
Absorption coefficient	$[\alpha] = \mathrm{m^{-1}}$	$[\alpha] = \mathrm{cm^{-1}}$
Intensity	$[I] = \mathrm{W/m^2}$	$[I] = \mathrm{erg/s/cm^2}$
Power	$[\mathcal{P}] = \mathrm{W}$	$[\mathcal{P}] = \mathrm{erg/s}$

at the input of the nonlinear medium at $z = 0$. The output intensities of all waves are then determined at the end of the nonlinear medium at $z = L$. Boundary effects are ignored. When there are two pump waves, it will be understood that $I_{p1} \cong I_{p2}$, and conversion efficiencies will be in terms of the smaller pump power.

A. Negligible Pump Depletion

As a first order of approximation, it is assumed that in the frequency conversion process there is negligible depletion of the pump beam(s) power. This means

Table 2 Conversion Formulas

$$P^{(2)} \quad (\mathrm{SI}) = \tfrac{1}{3} \times 10^{-5} P^{(2)} \quad (\mathrm{cgs})$$

$$E \quad (\mathrm{SI}) = 3 \times 10^{4} E \quad (\mathrm{cgs})$$

$$d \quad (\mathrm{SI}) = (4\pi/3) \times 10^{-4} d \quad (\mathrm{cgs})$$

$$L \quad (\mathrm{SI}) = 10^{-2} L \quad (\mathrm{cgs})$$

$$c \quad (\mathrm{SI}) = 10^{-2} c \quad (\mathrm{cgs})$$

$$\lambda \quad (\mathrm{SI}) = 10^{-2} \lambda \quad (\mathrm{cgs})$$

$$k \quad (\mathrm{SI}) = 10^{2} k \quad (\mathrm{cgs})$$

$$\alpha \quad (\mathrm{SI}) = 10^{2} \alpha \quad (\mathrm{cgs})$$

$$I \quad (\mathrm{SI}) = 10^{-3} I \quad (\mathrm{cgs})$$

$$\mathcal{P} \quad (\mathrm{SI}) = 10^{-7} \mathcal{P} \quad (\mathrm{cgs})$$

Table 3 Conversion Efficiency Formulas in the Infinite Plane Wave, Nondepleted Pump Approximation

SHG $(2\omega = \omega + \omega)$	$\eta_{2\omega} = \frac{\mathcal{P}_{2\omega}}{\mathcal{P}_{\omega}} = \eta_{2\omega}^{0} \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2}$ $\eta_{2\omega}^{0} = \frac{8\pi^2 d_{\text{eff}}^2 L^2 I_\omega}{\varepsilon_0 n_\omega^2 n_{2\omega} c \lambda_\omega^2}$ (SI) $\quad \eta_{2\omega}^{0} = \frac{512\pi^5 d_{\text{eff}}^2 L^2 I_\omega}{n_\omega^2 n_{2\omega} c \lambda_\omega^2}$ (cgs)
SFG $(\omega_s = \omega_{p1} + \omega_{p2})$	$\eta_s = \frac{\mathcal{P}_s}{\mathcal{P}_{p2}} = \eta_s^0 \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2}$ $\eta_s^0 = \frac{8\pi^2 d_{\text{eff}}^2 L^2 I_{p1}}{\varepsilon_0 n_{p1} n_{p2} n_s c \lambda_s^2}$ (SI) $\quad \eta_s^0 = \frac{512\pi^5 d_{\text{eff}}^2 L^2 I_{p1}}{n_{p1} n_{p2} n_s c \lambda_s^2}$ (cgs)
DFG $(\omega_d = \omega_{p1} - \omega_{p2})$	$\eta_d = \frac{\mathcal{P}_d}{\mathcal{P}_{p2}} = \eta_d^0 \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2}$ $\eta_d^0 = \frac{8\pi^2 d_{\text{eff}}^2 L^2 I_{p1}}{\varepsilon_0 n_{p1} n_{p2} n_d c \lambda_d^2}$ (SI) $\quad \eta_d^0 = \frac{512\pi^5 d_{\text{eff}}^2 L^2 I_{p1}}{n_{p1} n_{p2} n_d c \lambda_d^2}$ (cgs)

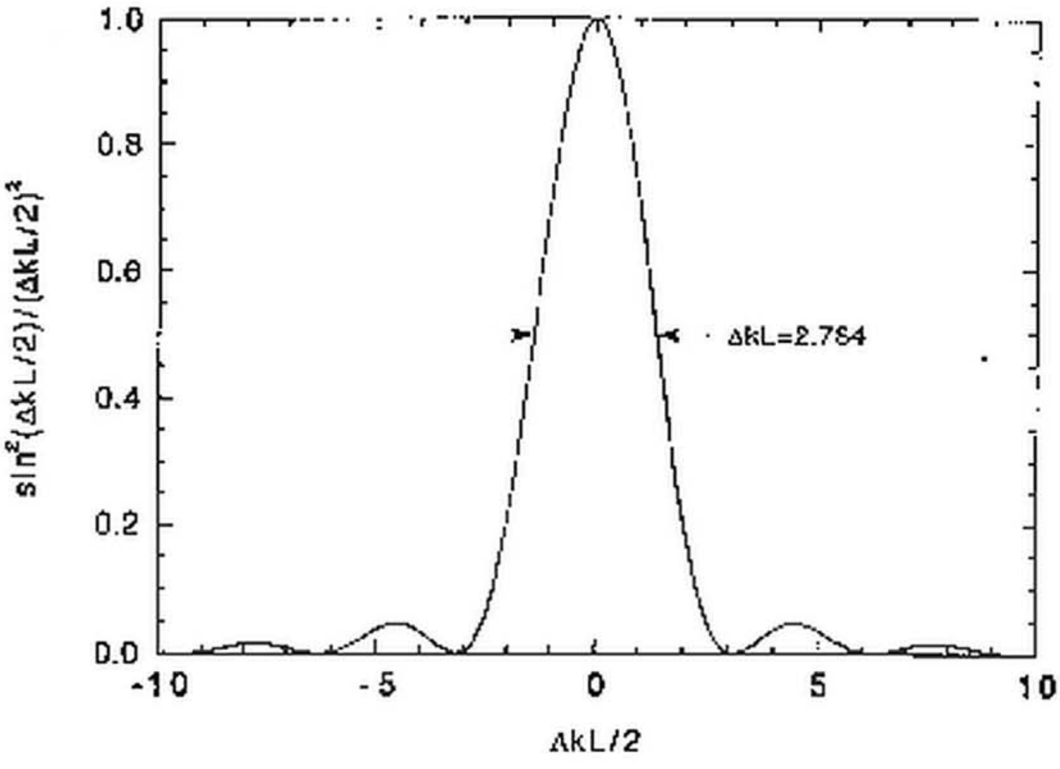

Figure 4 Plot of the sinc^2 function, which describes the effects of phase mismatch in frequency conversion processes.