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Fourth Order Nonlinear Intensity and the corresponding Refractive Index in Uniaxial Crystals

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Abstract

Nonlinear effects occur whenever the optical fields associated with one or more intense light such as from laser beams propagating in a crystal are large enough to produce polarization fields. This paper describes how the fourth order nonlinear intensity and the corresponding effective refractive index that is intensity dependent can be obtained using Maxwell's equations. Applications have been elucidated for some uniaxial crystals.

Keywords: Refractive index, Nonlinear effects, Maxwell's equations, Intensity and uniaxial crystals.

1. Introduction

Nonlinear optical phenomena occur typically at high optical intensities, or, equivalently, at high average photon number in an optical resonance [1]. The field of nonlinear optics as described by Cowan and Young [2] came into light in 1961 when the frequency doubling of a ruby laser was observed upon passing through a quartz crystal. Nonlinear optics concerns the response of matter to intense electromagnetic field such as the one obtained from laser light, in which the matter responds in a nonlinear manner to the incident radiation fields. The nonlinear response can result in intensity dependent variation of the propagation characteristics of the radiation fields that propagate at new frequencies or in new directions. Practical applications of nonlinear optical effects have risen as a direct consequence of the invention of lasers [3]. Nonlinear optics has played an increasing role in laser science, making it possible to generate coherent light more efficiently, and in spectral regions that cannot be directly accessed by laser [4]. The second harmonic generation has been shown to be a valuable detection tool in both industry and academics [5]. The induced polarization **P** in a medium and the electric field **E** of the electromagnetic wave propagating in the medium are related by [6]

$$P = \varepsilon_0 \chi E \qquad (1)$$

where χ is the dielectric susceptibility of the medium, that depends on the frequency, but independent of the field **E**. Equation (1) is valid for the field strengths of conventional source. With sufficiently intense laser radiation that is associated with THz, equation (1) does not hold well, and hence needs to be generalized [7]. The polarization induced in a medium by optical fields can be represented by a power series in the optical fields [8, 9]. The power series of equation (1) is therefore

$$P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \chi^{(4)} E^4 + \ldots) \qquad (2)$$

where $\chi^{(1)}$ is the linear susceptibility, and $\chi^{(2)}$, $\chi^{(3)}$, $\chi^{(4)}$ and so on are the nonlinear susceptibilities. The fourth order susceptibility $\chi^{(4)}$ is responsible for fourth harmonic generation. A medium which lacks inversion symmetry at the molecular level has non zero second (all even) order susceptibility [10]. Fischer, et. al. [11] puts it thus "for a material to exhibit a coherent second order nonlinear optical responses, it needs to be noncentrosymmetric on macroscopic scale" However, this work is only concerned with the fourth order, and therefore the power series expansion of equation (2) stops at the fourth order. If the field is low, as it is in the case of ordinary light sources, only the first term in equation (2) can be retained. Other optical characteristics of the medium such as dielectric permittivity, refractive index, e.t.c which depend upon susceptibility also become functions of the field strength **E** , when the field is high [7]. If the field incident on a medium is of

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the form

$$E = E_0 \cos \omega t \tag{3}$$

then, equation (2) becomes

$$P = \varepsilon_0 (\chi^{(1)} E_0 \cos \omega t + \chi^{(2)} E_0^2 \cos^2 \omega t + \chi^{(3)} E_0^2 \cos^3 \omega t + \chi^{(4)} E_0^4 \cos^4 \omega t + \ldots) \tag{4}$$

Using the following trigonometric functions

$$\cos^2 \omega t = \frac{1}{2}(1 + \cos 2\omega t); \quad \cos^3 \omega t = \frac{1}{4}(\cos 3\omega t + 3\cos \omega t) \text{ and}$$
$$\cos^4 \omega t = \frac{1}{4}\left(\frac{3}{4} + 2\cos 2\omega t + \frac{1}{2}\cos 4\omega t\right) \tag{5}$$

equation (2) becomes

$$P = \varepsilon_0 \left(\frac{1}{2}\chi^{(2)} + \frac{3}{8}\chi^{(4)} E_0^2\right) E_0^2 + \varepsilon_0 \left(\chi^{(1)} + \frac{3}{4}\chi^{(3)} E_0^2\right) E_0 \cos \omega t$$
$$+ \frac{1}{2}\varepsilon_0 \left(\chi^{(2)} E_0 + \chi^{(4)} E_0^3\right) E_0 \cos 2\omega t + \frac{1}{4}\varepsilon_0 \chi^{(3)} E_0^3 \cos 3\omega t$$
$$+ \frac{1}{4}\varepsilon_0 \chi^{(4)} E_0^4 \cos 4\omega t \tag{6}$$

The first term of equation (6) is a constant that gives the d.c field across the medium, and has comparatively little practical importance [7]. The second term is called the first or fundamental harmonic of polarization. The third term that oscillates at frequency 2ω is called the second harmonic generation of polarization. The third term that contains 3ω is called the third harmonic generation, while the fifth term that contains frequency 4ω is called the fourth harmonic generation.
The polarization **P(t)** under the influence of an applied electric field can be described in terms of power series as [6, 12]

$$P(t) = P^{(0)}(t) + P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + P^{(4)}(t) + \ldots \tag{7}$$

Comparing equations (6) and (7), the fourth order polarization can be written as

$$P^{(4\omega)}(t) = \frac{1}{4}\varepsilon_0 \chi^{(4)} E_0^4 \cos 4\omega t \tag{8}$$

Comparing equations (4), (6), (7) and (8), the fourth order electric field is

$$E^{(4\omega)}(t) = \frac{1}{4} E_0^4 \cos 4\omega t \tag{9}$$

In this work, the relationship between the output intensity and the fourth order nonlinear electric field is obtained based on the contribution of fourth harmonic generation at frequency 4ω.

Theory

Maxwell's laws govern the interaction of bodies which are magnetically or electrically charged or both. The bodies and their charges may either be stationary or in motion [13]. According to Ubachs, 2001 [14], light propagating through a medium or vacuum may be described by a transverse wave, where the oscillating electric and magnetic field components are solutions to the Maxwell's equations. Also, the nonlinear polarizations induced in the medium have to obey these equations. The understanding of laser requires some knowledge of the way in which light and matter interact. The Maxwell's equations provide the most fundamental description of electric and magnetic fields. For a neutral dielectric medium (one with no free charges), the four Maxwell's equations are;

$$\nabla . \mathbf{D} = 0 \tag{10}$$
$$\nabla . \mathbf{B} = 0 \tag{11}$$
$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{12}$$
$$\nabla \times \mathbf{H} = -\frac{\partial \mathbf{D}}{\partial t} \tag{13}$$

where **E** is the electric field in N/C, **B** is the magnetic field in Tesla, **D** is the electric displacement in C/m^2, and **H** is the magnetic intensity in Am^{-1}. Duffin, 1981, [15] defines the electric displacement as

$$\mathbf{D} = \varepsilon_o \mathbf{E} + \mathbf{P} \tag{14}$$

where ε_o is the permittivity of free space (~ 8.85x10^{-12} C^2/N.m^2), **P** is the polarization in C/m^2, and gives the dielectric dipole moment per unit volume of the medium. **P** is the only term in the Maxwell's equations relating directly to the medium. From the fact that

$$\varepsilon_o \mu_o = \frac{1}{c^2} \tag{15}$$

where c is the speed of light (~ 3 x 10^8 m/s), equations (12), (14) and (15), lead to

$$\nabla^2 \mathbf{E} - \nabla(\nabla.\mathbf{E}) - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\varepsilon_o c^2}\frac{\partial^2 \mathbf{P}}{\partial t^2} \tag{16}$$

Equation (16) is a partial differential equation with independent variables x, y, z and t. It tells us how the electric field **E** depends on the electric dipole moment density **P** of the medium. If one considers the transverse fields (radiation fields) of which the THz radiation is an example, then

$$\nabla.\mathbf{E} = 0 \tag{17}$$

Transverse fields therefore satisfy the inhomogeneous wave equation

$$\nabla^2 \mathbf{E} - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\varepsilon_o c^2}\frac{\partial^2 \mathbf{P}}{\partial t^2} \tag{18}$$

Equation (18) is the fundamental electromagnetic wave equation.

Methodology

The method employed in this work involves the use of fourth order nonlinear induced polarization $\mathbf{P}^{(4)}(\mathbf{t})$ equation. Since transverse waves satisfy the inhomogeneous wave equation (16), equation (18) can be written as

$$\nabla^2 E - \varepsilon_o \mu_o \frac{\partial^2 E}{\partial t^2} = \mu_o \frac{\partial^2 P}{\partial t^2} \tag{19}$$

while the corresponding fourth harmonic field will thus be written as

$$E = \frac{1}{2}\left[E_{4\omega}(z)e^{-i(4\omega t - k_{4\omega}z)} + E_{4\omega}^*(z)e^{i(4\omega t - k_{4\omega}z)}\right] \tag{20}$$

Lekner, 1993, [17] defines $k = n\omega/c$. From equation (20) therefore, $k_{4\omega} = 4\omega\, n(4\omega)/c$ and $n(4\omega) = (\varepsilon_{4\omega}/\varepsilon_0)^{1/2}$ is the refractive index of the medium for radiation of frequency 4ω. Using equations (19) and (20), one gets

$$\nabla^2 E = \frac{\partial^2 E}{\partial z^2} = \frac{1}{2}\left[\{4ik_{4\omega}\frac{dE_{4\omega}}{dz} - k_{4\omega}^2 E_{4\omega}\}e^{-i(4\omega t - k_{4\omega}z)} + \{-4ik_{4\omega}\frac{dE_{4\omega}^*}{dz} - k_{4\omega}^2 E_{4\omega}^*\}e^{i(4\omega t - k_{4\omega}z)}\right] \tag{21}$$

$$\frac{\partial^2 E}{\partial t^2} = -8\omega^2\left[E_{4\omega}(z)e^{-i(4\omega t - k_{4\omega}z)} + E_{4\omega}^*(z)e^{i(4\omega t - k_{4\omega}z)}\right] \tag{22}$$

Therefore

$$\nabla E - \varepsilon_0 \mu_0 \frac{\partial^2 E}{\partial t^2} = \left[2ik_{4\omega}\frac{dE_{4\omega}}{dz} - \frac{1}{2}\left(k_{4\omega}^2 - 16\varepsilon_0\mu_0\omega^2\right)E_{4\omega}\right]e^{-i(4\omega t - k_\omega z)} + complex\ conjugate \tag{23}$$

Note that the right hand side of equation (19) has both the linear and the nonlinear contribution at frequency 4ω, such that

$$P = \frac{1}{2}\left[P_{4\omega}^{(L)}e^{-i(4\omega t - k_{4\omega}z)} + P_{(4\omega)}^{(L)*}e^{i(4\omega t - k_{4\omega}z)}\right] + \frac{1}{2}\left[P_{4\omega}^{(NL)}e^{-4i(\omega t - k_\omega z)} + P_{4\omega}^{(NL)*}e^{4i(\omega t - k_\omega z)}\right] \tag{24}$$

so that

$$\mu_o \frac{\partial^2 P}{\partial t^2} = -8\mu_o\omega^2 P_{4\omega}^{(L)}e^{-i(4\omega t - k_{4\omega}z)} - 8\mu_o\omega^2 P_{4\omega}^{(NL)}e^{-4i(\omega t - k_\omega z)} + complex\ conjugate \tag{25}$$

From equations (23) and (25), one obtains

$$\left[2ik_{4\omega}\frac{dE_{4\omega}}{dz}-\frac{1}{2}(k_{4\omega}^{2}-16\varepsilon_{o}\mu_{o}\omega^{2})E_{4\omega}\right]e^{-i(4\omega t-k_{4\omega}z)}=$$
$$-8\mu_{o}\omega^{2}P_{4\omega}^{(L)}e^{-i(4\omega t-k_{4\omega}z)}-8\mu_{o}\omega^{2}P_{4\omega}^{(NL)}e^{-4i(\omega t-k_{\omega}z)} \quad (26)$$

But $P=\varepsilon_{o}\chi E$, and, at frequency 4ω, P becomes

$$P_{4\omega}^{L}=\varepsilon_{o}\chi(4\omega)E_{4\omega} \quad (27)$$

From Appendix (B3)

$$k_{4\omega}^{2}=16\varepsilon_{o}\mu_{o}\omega^{2}[1+\chi(4\omega)] \quad (28)$$

where $\chi(4\omega)$ is the susceptibility for the frequency 4ω. Substituting equations (27) and (28) in (26), one gets

$$2ik_{4\omega}\frac{dE_{4\omega}}{dz}e^{-i(4\omega t-k_{4\omega}z)}-8\mu_{o}\varepsilon_{o}\omega^{2}E_{4\omega}e^{-i(4\omega t-k_{4\omega}z)}-8\omega^{2}\varepsilon_{o}\mu_{o}\chi(4\omega)E_{4\omega}e^{-i(4\omega t-k_{4\omega}z)}$$
$$+8\varepsilon_{o}\mu_{o}\omega^{2}E_{4\omega}e^{-i(4\omega t-k_{4\omega}z)}=-8\mu_{o}\varepsilon_{o}\omega^{2}\chi(4\omega)E_{4\omega}e^{-i(4\omega t-k_{4\omega}z)}-8\mu_{o}\omega^{2}P_{4\omega}^{(NL)}e^{-4i(\omega t-k_{\omega}z)} \quad (29)$$

From equation (29)

$$\frac{dE_{4\omega}}{dz}=\frac{4i\mu_{0}\omega^{2}}{k_{4\omega}}P_{4\omega}^{(NL)}e^{i(4k_{\omega}-k_{4\omega})z} \quad (30)$$

From Appendix (B3) and equation (30),

$$\frac{dE_{4\omega}}{dz}=i\omega\sqrt{\frac{\mu_{o}}{\varepsilon_{4\omega}}}.P_{4\omega}^{(NL)}e^{i(4k_{\omega}-k_{4\omega})z} \quad (31)$$

Equation 31 gives the relationship between fourth harmonic field $E_{4\omega}$ and the nonlinear polarization $P_{4\omega}^{(NL)}$ From Appendix (A15)

$$P_{4\omega}^{(NL)}=gE_{\omega}^{4}(z) \quad (32)$$

where g is called the electronic polarizability. Equation (31) can therefore be written as

$$\frac{dE_{4\omega}}{dz}=i\omega\sqrt{\frac{\mu_{o}}{\varepsilon_{2\omega}}}.gE_{\omega}^{4}(z)e^{i\Delta kz} \quad (33)$$

where $\Delta k=4k_{\omega}-k_{4\omega}$

For a special and a simple case, it can be assumed that there is a little attenuation of the fundamental wave, such that E_{ω} can be considered constant, such that $E_{\omega}(z)\approx E_{\omega}(0)$, so that equation (33) becomes

$$E_{4\omega}(z)\approx i\omega\sqrt{\frac{\mu_{o}}{\varepsilon_{2\omega}}}\,gE_{\omega}^{4}(0)\left(\frac{e^{i\Delta kz}-1}{i\Delta k}\right) \quad (34)$$

Using Appendix (C1), leads to

$$E_{4\omega}(z)=i\omega\sqrt{\frac{\mu_{o}}{\varepsilon_{2\omega}}}\,gE_{\omega}^{4}(0)z\exp\left(\frac{i\Delta kL}{2}\right)\left(\frac{\sin\frac{1}{2}\Delta kz}{\frac{1}{2}\Delta kz}\right) \quad (35)$$

If the nonlinear crystal is of length L, and the fourth harmonic field is at the exit face of the crystal such that z = L, equation (35) becomes

$$E_{4\omega}(L)=i\omega\sqrt{\frac{\mu_{0}}{\varepsilon_{4\omega}}}gE_{\omega}^{4}(0)L\exp\left(\frac{i\Delta kL}{2}\right)\left(\frac{\sin\frac{1}{2}\Delta kL}{\frac{1}{2}\Delta kL}\right) \quad (36)$$

For a plane wave represented by equation (3), the intensity *I* is defined as [12]

$$I(\omega)=\frac{1}{2}\varepsilon_o c n_l |E|^2 \qquad (37)$$

For a plane wave moving with a frequency 4ω of the fundamental, the intensity I is

$$I_{4\omega}(\omega)=\frac{1}{2}\varepsilon_o c n(4\omega)|E_{4\omega}|^2 \qquad (38)$$

using $c=\frac{1}{\sqrt{\mu_o\varepsilon_o}}$ and $n^2(4\omega)\varepsilon_o=\varepsilon_{4\omega}$ in equations (36) and (38)

$$I_{4\omega}(\omega)=E_{(4\omega)}(L)E_{(4\omega)}(L)^*=\frac{\omega^2 g^2}{n(4\omega)}\left(\frac{\mu_o}{\varepsilon_o}\right)^{1/2}|E(0)|^8 L^2\frac{\sin^2\left(\frac{1}{2}\Delta kz\right)}{\left(\frac{1}{2}\Delta kz\right)^2} \qquad (39)$$

Equation (39) gives the intensity of fourth harmonic generation associated with fourth order nonlinear electric field. From equation (6), $\chi^{(3)}$ vanishes for uniaxial crystals. The polarization can thus be written as

$$P=\varepsilon_0\chi^{(1)}E_0\cos\omega t+\frac{1}{2}\varepsilon_0\chi^{(2)}E_0E_o\cos 2\omega t+\frac{1}{2}\varepsilon_0\chi^{(4)}E_0^3E_o\cos 2\omega t+\frac{1}{4}\varepsilon_0\chi^{(4)}E_0^3E_o\cos 4\omega t \qquad (40)$$

Since this work is restricted to fourth order nonlinearity, higher order terms greater than four are neglected. Due to the variations of optical nonlinear properties along the axes of the crystal, there is phase mismatch between frequencies ω, 2ω and 4ω of equation (40). In odd order nonlinearity, the phase matching condition is easily obtained [10]. In equation (40), the phase matching condition between frequencies ω and 2ω can be satisfied by chosen a direction in the uniaxial crystal such that the second harmonic propagates as an ordinary wave and the fundamental as an extraordinary wave (or vice versa) with equal velocity (hence equal indices of refraction as well as frequency) [6]. For negative uniaxial crystals, the diagram to illustrate this phase matching condition is shown in Fig.1.

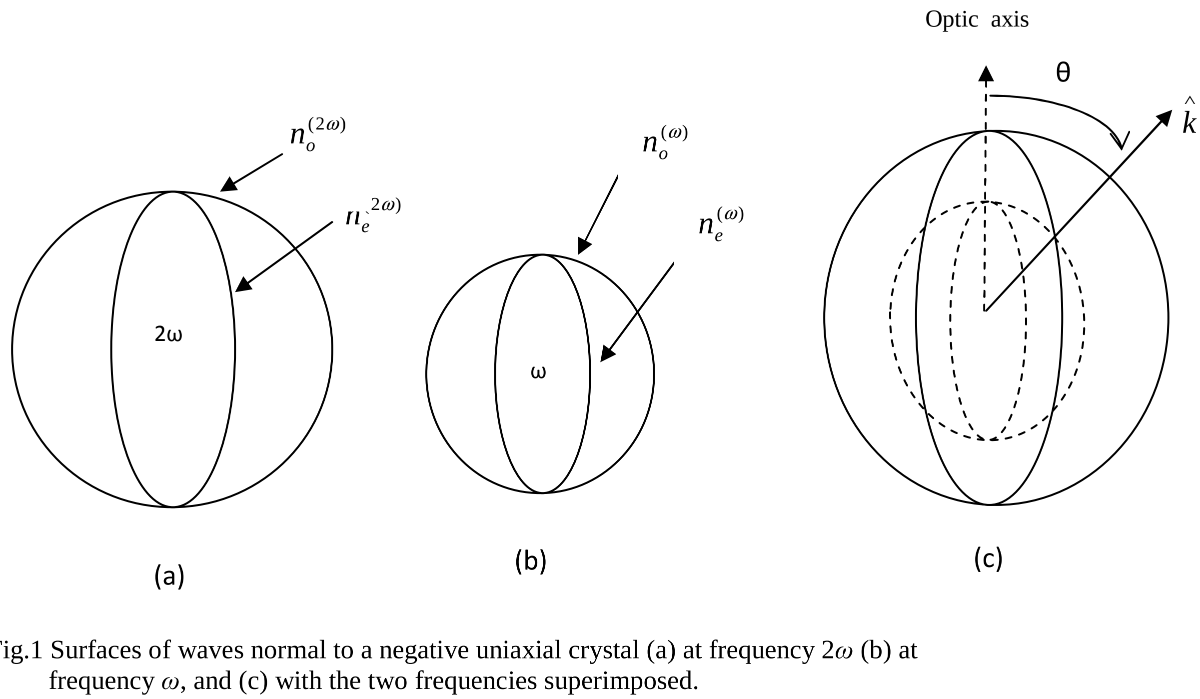

Fig.1 Surfaces of waves normal to a negative uniaxial crystal (a) at frequency 2ω (b) at frequency ω, and (c) with the two frequencies superimposed.

From Fig.1 (c), the two waves at frequencies ω and 2ω are superimposed in the direction $\hat{k}$. The spherical surface of radius $n_o(\omega)$ of the ordinary wave of frequency ω intersects the ellipsoid index $n_e^{(2\omega)}(\theta)$ (where θ is the angle between the optic axis and $\hat{k}$ direction) of the extraordinary wave of frequency 2ω. At the point of intersection, $n_o^{(\omega)} = n_e^{(2\omega)}(\theta)$, thus satisfying the phase matching condition of equation (40). This stated phase matching condition is also true for positive uniaxial crystals. Equation (40) can thus be written as

$$P = \varepsilon_0 \chi^{(1)} E_0 \cos \omega t + \frac{1}{2} \varepsilon_0 \chi^{(2)} E_0 E_o \cos \omega t + \frac{1}{2} \varepsilon_0 \chi^{(4)} E_0^3 E_o \cos \omega t \tag{41}$$

the first term of equation (41) is the linear term, while the second and the third terms are nonlinear terms of the polarization. For a plane wave represented by equation (3), the intensity is defined as

$$I = \tfrac{1}{2} c \varepsilon_o n_l E_o^2 \tag{42}$$

where n_l is the linear refractive index of the crystal at low fields. Using equations (3), (41) and (42), the polarization can be written as

$$P = \varepsilon_o \chi^{(1)} E + \frac{\chi^{(2)} E}{c n_l E_o} I + \frac{2 \chi^{(4)} E_o E}{c^2 n_l^2 \varepsilon_o^2} I^2 \tag{43}$$

Singh and Singh, 2007 [10] defined the effective susceptibility (χ_{eff}) of a medium as

$$\chi_{eff} = \frac{P}{\varepsilon_o E} \tag{44}$$

Thus, from equations (43) and (44)

$$\chi_{eff} = \chi^{(1)} + \frac{\chi^{(2)}}{c n_l \varepsilon_o E_o} I + \frac{2 \chi^{(4)} E_o}{c^2 n_l^2 \varepsilon_o^2} I^2 \tag{45}$$

The effective refractive index (n_{eff}) is related to the effective susceptibility as

$$n_{eff} = (1 + \chi_{eff})^{1/2} \equiv \left(1 + \chi^{(1)} + \frac{\chi^{(2)}}{c n_i \varepsilon_0 E_0} I + \frac{2 \chi^{(4)} E_0}{c^2 n_i^2 \varepsilon_0^2} I^2 \right)^{1/2} \tag{46}$$

Equation (46) can be said to contain both the linear and the nonlinear terms, such that it can be written as

$$n_{eff} = (\chi_l + \chi_{nl})^{\frac{1}{2}} \tag{47}$$

Equation (47) can further be approximated using Taylor's series expansion and can be written as

$$n_{eff} = \left(\chi_l + \frac{1}{2} \chi_{nl} \right) \tag{48}$$

such that

$$\chi_l = (1 + \chi^{(1)}) \tag{49}$$

and

$$\chi_{nl} = \frac{\chi^{(2)}}{2 c n_l \varepsilon_o E_o} I + \frac{\chi^{(4)} E_o}{c^2 n_l^2 \varepsilon_o^2} I^2 \tag{50}$$

The effective refractive index that is intensity dependent for fourth order nonlinearity can thus be written as

$$n_{eff} = 1 + \chi^{(1)} + \frac{\chi^{(2)}}{2 c n_l \varepsilon_o E_o} I + \frac{\chi^{(4)} E_o}{c^2 n_l^2 \varepsilon_o^2} I^2 \tag{51}$$

Conclusions

If equation 51 should only be written in terms of the refractive indices and electromagnetic intensity, then, we have

$$n_{eff} = n_0 + n_2 I + n_4 I^2 \tag{52}$$

where

$$n_0 = 1 + \chi^{(1)} \tag{53a}$$

$$n_2 = \frac{\chi^{(2)}}{2c\, n_l \varepsilon_o E_o} \tag{53b}$$

$$n_4 = \frac{\chi^{(4)} E_o}{c^2 n_l^2 \varepsilon_o^2} \tag{53c}$$

n_0 is the linear refractive index, n_2 is the second order nonlinear refractive index, and n_4 is the forth order nonlinear refractive index. The result in equation (52) is similar to what Miller, et. al., 1979 [18] obtained for second order effective refractive index given as

$$n_{eff} = n_0 + n_2 I \tag{53 }$$

Similarly, the expression for the intensity obtained in equation (39) is comparable with what Ubachs, 2001 [14] defined as the output intensity for second harmonics, given as

$$I_{2\omega}(E) = E_{(2\omega)}(L) E_{(2\omega)}(L)^* = \frac{\omega^2 d^2}{n^2(2\omega)}\left(\frac{\mu_o}{\varepsilon_o}\right)\left|E(0)\right|^4 L^2 \frac{\sin^2\left(\frac{1}{2}\Delta kz\right)}{\left(\frac{1}{2}\Delta kz\right)^2} \tag{54}$$

This work has established the relationship between fourth order intensity $I_{4\omega}(E)$ and fourth order nonlinear electric field $\mathbf{E}_{(4\omega)}$. The result shows that the fourth order intensity is proportional to the eighth power of the corresponding nonlinear electric field. Using the stated phase matching conditions, the effective refractive index is certainly intensity dependent.

Appendix A

Fourth Order Electronic Polarizability g(ω)

Consider the nonlinear electron oscillator equation

$$\frac{d^2 x}{dt^2} + \omega_o^2 x + ax^2 = \frac{e}{m} E_o \cos \omega t \tag{A1}$$

If the nonlinear term ax^2 is considered to be small, then equation (A1) can have an approximation $x^{(1)}t$ given by [12]

$$\frac{d^2 x^{(1)}(t)}{dt^2} + \omega_o^2 x^{(1)}(t) = \frac{e}{m} E_o \cos \omega t \tag{A2}$$

Equation (A2) has a solution of the form

$$x^{(1)}(t) = \frac{e}{m\left(\omega_o^2 - \omega^2\right)} E_o \cos \omega t \tag{A3}$$

From equation (A1), a better approximation to $x(t)$, labelled $x^{(2)}(t)$ is given as

$$\frac{d^2 x^{(2)}(t)}{dt^2} + \omega_o^2 x^{(2)}(t) = \frac{e}{m} E_o \cos \omega t - a\left[x^{(1)}(t)\right]^2 \tag{A4}$$

In equation (A4), the effect of the nonlinear term is included. From (A3)

$$\left[x^{(1)}(t)\right]^2 = \frac{1}{2}\left(\frac{e}{m(\omega_o^2 - \omega^2)}\right)^2 E_o^2 (1 + \cos 2\omega t) \tag{A5}$$

Substituting equation (A5) in (A4) yields

$$\frac{d^2x^{(2)}(t)}{dt^2}+\omega_o^2x^{(2)}(t)=\frac{e}{m}E_o\cos\omega t-\frac{a}{2}\left(\frac{e}{m(\omega_o^2-\omega^2)}\right)E_o^2-\frac{a}{2}\left(\frac{e}{m(\omega_o^2-\omega^2)}\right)^2E_o^2\cos 2\omega t \quad \text{(A6)}$$

Equation (A6) has a steadily driven solution given as

$$x^{(2)}(t)=\frac{e}{m(\omega_o^2-\omega^2)}E_o\cos\omega t-\frac{a}{2\omega_o^2}\left(\frac{e}{m(\omega_o^2-\omega^2)}\right)^2E_o^2$$
$$-\frac{a}{2}\cdot\frac{1}{(\omega_o^2-4\omega^2)}\left(\frac{e}{m(\omega_o^2-\omega^2)}\right)^2E_o^2\cos 2\omega t \quad \text{(A7)}$$

Equation (A7) shows that the improved (second) approximation to $x(t)$ has a term oscillating at fundamental driving field frequency ω (first term on R.H.S), the d.c or static term (second term on R.H.S) and another term oscillating at second harmonic frequency 2ω. Equation (A7) can be written as

$$x^{(2)}(t)=x_o+x_\omega+x_{2\omega} \quad \text{(A8)}$$

Such that

$$x_o=-\frac{a}{2\omega_o^2}\left(\frac{e}{m(\omega_o^2-\omega^2)}\right)^2|E_\omega|^2 \quad \text{(A9)}$$

$$x_\omega=\frac{e}{m(\omega_o^2-\omega^2)}E_\omega \quad \text{(A10)}$$

$$x_{2\omega}=-\frac{a}{2}\frac{1}{(\omega_o^2-4\omega^2)}\left(\frac{e}{m(\omega_o^2-\omega^2)}\right)^2|E_o|^2 \quad \text{(A11)}$$

where x_o is the d.c or static electron displacement; x_ω is the electron displacement at ω, and $x_{2\omega}$ is the electron displacement at 2ω. But the polarization density is given as

$$P=Nex \quad \text{(A12)}$$

where N is the electron density in m^{-3}, and e is the electronic charge. For second harmonics, the nonlinear term of (A12) is given as

$$P_{2\omega}^{(NL)}=\frac{-Nae^3}{2m^2(\omega_o^2-4\omega^2)(\omega_o^2-\omega^2)^2}E_\omega^2(z) \quad \text{(A13)}$$

One can define a quantity d such that (A13) becomes [12]

$$P_{2\omega}^{(NL)}=dE_\omega^2(z) \quad \text{(A14)}$$

where

$$d(\omega)=\frac{-Nae^3}{2m^2(\omega_o^2-4\omega^2)(\omega_o^2-\omega^2)^2}$$

and is called the electronic polarizability. According to Ubachs, 2001[14], the electronic polarizability associated with fourth harmonics is of the same order as the product of three second order electronic polarizabilities. Hence (A14) can be written in terms of the fourth order as

$$P_{4\omega}^{(NL)}=d^3E_\omega^4\equiv gE_\omega^4(z) \quad \text{(A15)}$$

where g is the electronic polarizability associated with fourth order harmonics. Equation (A15) has been used as equation (32).

Appendix B

Fourth Order Wave Number ($k_{4\omega}$) and Refractive index ($n^2(4\omega)$)
According to Milloni and Eberly (1988), [9], *k* must satisfy the dispersion relation

$$k^2 = \frac{\omega^2}{c^2}\left(1 + \frac{N\alpha(\omega)}{\varepsilon_o}\right) = \frac{\omega^2}{c^2} n^2(\omega)$$

$$or \qquad \text{(B1)}$$

$$k = \frac{n(\omega)\,\omega}{c}$$

For fourth harmonic generation, and using $c = \frac{1}{\sqrt{\mu_o \varepsilon_o}}$ and $n^2(4\omega)\varepsilon_o = \varepsilon_{4\omega}$ B1 becomes

$$k_{4\omega} = \frac{n(4\omega)\,4\omega}{c}$$

$$= \left(\frac{\varepsilon_{4\omega}}{\varepsilon_o}\right)^{1/2} .(\mu_o \varepsilon_o)^{1/2} .4\omega$$

$$= (\varepsilon_{4\omega}\mu_o)^{1/2} ,4\omega \qquad \text{(B2)}$$

Equation (B2) is used in equation (31). From (B2),

$$k_{4\omega}^2 = (4\omega)^2 \varepsilon_{4\omega}\mu_0 \equiv 16\omega^2 \varepsilon_0 \mu_0 n^2(4\omega) = 16\omega^2 \varepsilon_0 \mu_0 [1 + \chi(4\omega)] \qquad \text{(B3)}$$

for $\quad n^2(4\omega) = 1 + \chi(4\omega)$

Equation (B3) is used as equation (29).

Appendix C

Using standard integral for exponential functions

$$\int \frac{dx}{e^{ax}} = \int e^{-ax} dx = -\frac{1}{ae^{ax}} = -\frac{1}{a} e^{-ax}$$

$$\int_0^z e^{i\Delta kz'} dz' = \frac{1}{i\Delta k}\left[e^{i\Delta kz'}\right]_0^z$$

$$= \frac{1}{i\Delta k}\left[e^{i\Delta kz} - 1\right]$$

$$= \frac{e^{i\Delta kz/2}}{i\Delta k}\left[e^{i\Delta kz/2} - e^{-i\Delta kz/2}\right] = ze^{i\Delta kz/2}\left[\frac{\sin \frac{1}{2}\Delta kz}{\frac{1}{2}\Delta kz}\right] \qquad \text{(C1)}$$

where Euler's identities $e^{i\theta} = \cos\theta + i\sin\theta \quad and \quad e^{-i\theta} = \cos\theta - i\sin\theta$ have been used to get (C1), which is used in equation (35).

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