

Second-Harmonic Generation

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Abstract

The basic principles underlying the generation of a second-harmonic wave in dielectric materials were studied. Solving Maxwell's equations, it was discovered theoretically that when light with high intensity is incident on a nonlinear crystal, then a wave doubling the frequency of the incident light called the second-harmonic wave can be obtained by proper phase matching the fundamental and the second-harmonic waves. The intensity of the second-harmonic wave was shown to increase as the square of the intensity of the fundamental field under the approximation that the fundamental field was undepleted in the medium. However, experimental results showed that, due to the depletion of the fundamental wave as it passes through the medium, the increase of the intensity of the second-harmonic signal as the square of the fundamental wave reaches a saturation point, from which the intensity of the second-harmonic wave maintains a constant value.

Contents

Abstract	i
List of Figures	iv
1 General Background	1
1.1 Introduction	1
1.2 Linear Polarisation of a Dielectric Medium	1
1.3 Electron Oscillation Model and Linear Polarisation	2
1.4 Wave Equation in a Medium and Linear Polarisation	3
1.5 Linear Polarisation and Refractive Index	5
1.6 Relationship between Energy and Momentum of Photons	8
1.7 Origin of Nonlinear Polarisation	9
2 Second Order Nonlinear Optical Phenomena	13
2.1 Nonlinear Polarisation	13
2.1.1 Nonlinear Wave Equation	13
2.2 Second Order Nonlinear Optical Processes	15
2.2.1 Second-Harmonic Generation	15
2.2.2 Optical Sum Frequency Generation	17
2.2.3 Optical Difference-Frequency Generation	18
2.2.4 Nonlinear Crystals for Second-harmonic generation	19
3 Second-Harmonic Generation	20
3.1 Intensity of a Second-Harmonic Wave	20
3.2 Phase Matching Condition	25
3.2.1 Birefringence	27
3.2.2 Angle Phase Matching Method	27
4 Conclusion	30

A	Code for the computation of the variation of the refractive index with frequency	31
A.1	A code for the dispersion curve	31
A.2	A code for the imaginary part of the refractive index	31
B	Code to Compute Reduction in the Second Harmonic Generation and Coherence Length	33
B.1	Coherence length	33
C	Code for plotting the Intensity of the Second-Harmonic Signal	34
C.1	Fitted Quadratic Polynomial on the Generation of the Second-Harmonic Signal .	34
	Bibliography	37

List of Figures

1.1	Imaginary part of the refractive index versus frequency, $\omega_0 = 20$	7
1.2	Real part of the refractive index versus frequency $\omega_0 = 20$	7
2.1	Energy level description of second-harmonic generation from a quantum standpoint.	16
2.2	Energy level description of sum-frequency generation from a quantum standpoint.	18
2.3	Energy level description of difference-frequency generation form a quantum standpoint.	19
3.1	Intensity of the second-harmonic signal versus the average input power	24
3.2	Schematic experimental arrangement for detection of second-harmonic generation, where ω is the frequency of the incident laser beam.	25
3.3	The graph of $f(L) = \frac{(\sin^2 \alpha L)}{(\alpha L)^2}$, where $\alpha \equiv \Delta k/2$	26
3.4	Geomety for angle phase matching of second-harmonic generation.	28

1. General Background

1.1 Introduction

Laser physics and nonlinear optics are fields which have been intimately connected from their beginning[1]. Nonlinear optics, includes frequency conversions, and these new frequency generation processes has found its applications in different research areas including spectroscopy. Preceding the invention of lasers, the optical effects observed in materials as light passes through it has been treated in a linear manner, but with the invention of laser light with high intensities, interesting nonlinear optical effects have now been observed and studied. Nonlinear optics is the study of the phenomena that occur as a consequence of the enhancement of the optical properties of a material system by the presence of light with high intensity. Typically, only laser light is sufficiently intense to bring to the fore the nonlinear optical properties of a material. In fact, the beginning of the field of nonlinear optics is often taken to be the discovery of second-harmonic generation by Franken et al in 1961[2].

The phenomena are nonlinear in the sense that the response of a material to an applied optical field depends in a nonlinear manner upon the strength of the incident electric field. For example, second-harmonic generation occurs as a result of the part of the atomic response that depends on the square of the strength of the applied field. Consequently, the intensity of the light generated at the second-harmonic frequency tends to increase as the square of the intensity of the applied laser light[3]. This effect is discussed in detail in Section 3.1. In essence, we mean that for second-harmonic generation if we send in a laser light of about 694 nm we can observe an output light of double the frequency and so half the wavelength of the light at 347 nm. The basic questions then are; why are we able to observe this and how can this be efficiently generated? This work tries to explain the conditions under which this effect can be observed and how second-harmonic generation can be efficiently generated. Since second-harmonic generation is a nonlinear optical process, we will need to first understand how the linear process works, that is what we will explain in Chapter 1, the linear response of materials to an applied field and introduce the nonlinear effects. In Chapter 2 we will explain the nonlinear wave Equation and briefly explain the different second order nonlinear processes. In Chapter 3 we will explain how the efficiency of the second- harmonic generation is maximised and conclude this work with Chapter 4.

1.2 Linear Polarisation of a Dielectric Medium

Consider a dielectric medium (non-conducting) whose properties are isotropic. Application of a field to such a medium causes charge displacement, in which the negative charge distribution bound to the nuclei shifts in a direction opposite to the electric field while the positive end of the dipole is in the direction of the field . The dipole moment \mathbf{p} due to each atom is given by the product of the displaced charge q and the effective separation \mathbf{r} of the positive and negative charge of the atomic dipole

$$\mathbf{p} = -q\mathbf{r}.$$

The polarisation \mathbf{P} of the medium is then said to be the collective dipole moment per unit volume, given by

$$\mathbf{P} = -Ner, \quad (1.1)$$

where N is the number of elementary particles per volume, e is the magnitude of the electronic charge and \mathbf{r} is a vector representing the distance the charge $-e$ is displaced from its equilibrium position.

1.3 Electron Oscillation Model and Linear Polarisation

Let us assume that electrons behave as though the forces binding them to the nuclei are elastic forces described by Hooke's law, this means we are considering a linear response, where the restoring force is proportional to the displacement and oppositely directed. We can use this idea to obtain an expression for the polarisation in terms of the frequency of the applied field. The force on the electron due to the electric field is

$$\mathbf{F} = -e\mathbf{E} = -K\mathbf{r}, \quad (1.2)$$

where K is the restoring force constant. In Section 1.7 we shall consider additional nonlinear terms. Equation (1.2) leads to the formula for the static polarisation.

$$\mathbf{P} = \frac{Ne^2\mathbf{E}}{K}. \quad (1.3)$$

If the applied field varies with time, then Equation (1.3) no more holds. Using Newton's second law the Equation of motion of the localised charges now becomes

$$\mathbf{F} = m\frac{d^2\mathbf{r}}{dt^2} = (-K\mathbf{r} - e\mathbf{E}), \quad (1.4)$$

which can be written as

$$m\frac{d^2\mathbf{r}}{dt^2} + m\gamma\frac{d\mathbf{r}}{dt} + K\mathbf{r} = -e\mathbf{E}. \quad (1.5)$$

Here, $m\gamma\frac{d\mathbf{r}}{dt}$ was included and is the frictional damping term. The frictional damping term is proportional to the velocity of the electrons in the medium, and γ is a damping coefficient that measures the polarisation decay rate. Since the electric field is time dependent and is usually of the form $\mathbf{E} = \mathbf{E}_0e^{-i\omega t}$, we will assume that the time dependence of the electron motion has a solution of the form $\mathbf{r} = \mathbf{r}_0e^{-i\omega t}$, here ω is the angular oscillation frequency. Equation (1.5) leads to an Equation of the form

$$(-m\omega^2 - i\omega m\gamma + K)\mathbf{r} = -e\mathbf{E}. \quad (1.6)$$

Solving for \mathbf{r} we get

$$\mathbf{r} = \frac{-e\mathbf{E}}{(-m\omega^2 - i\omega m\gamma + K)} \quad (1.7)$$

and substituting in (1.1), we get a value for \mathbf{P} as

$$\mathbf{P} = \mathbf{P}(\omega) = \left(\frac{Ne^2}{-m\omega^2 - i\omega m\gamma + K} \right) \mathbf{E}. \quad (1.8)$$

This solution gives the static value for \mathbf{P} at $\omega = 0$ as would be expected and this also shows that the response of the polarisation to the applied electric field is frequency dependent. Dividing the numerator and the denominator of the right-hand side of Equation (1.8) by m and defining a resonant angular frequency ω_0 and oscillating frequency ν_0 as

$$\omega_0 \equiv \left(\frac{K}{m} \right)^{\frac{1}{2}} = 2\pi\nu_0, \quad (1.9)$$

\mathbf{P} then transforms to

$$\mathbf{P} = \left(\frac{Ne^2/m}{\omega_0^2 - \omega^2 - i\omega\gamma} \right) \mathbf{E}. \quad (1.10)$$

The resonant frequency ω_0 corresponds to that of a simple harmonic oscillator, and is strictly associated with the properties of the material and not of the electric field interacting with the material. Electrons are most easily perturbed or excited at the resonant frequency. The quantity in the bracket is referred to as the *linear susceptibility* of the material, often represented as χ , and is a property of the material since it enhances the linear polarisation \mathbf{P} when resonance is achieved. The linear polarisation of the material is then expressed as

$$\mathbf{P} = \epsilon_0\chi\mathbf{E}. \quad (1.11)$$

1.4 Wave Equation in a Medium and Linear Polarisation

In this Section we will derive the wave Equation for the linear polarisation of a medium, and we will also look at the nature of the wave vector \mathbf{k} propagating in the medium.

When we apply Maxwell's Equations to non-magnetic materials, and taking the magnetisation $\mathbf{M} = 0$, Maxwell's Equations then become[2]

$$\nabla \cdot \mathbf{E} = -\frac{1}{\epsilon_0} \nabla \cdot \mathbf{P}, \quad (1.12)$$

$$\nabla \cdot \mathbf{H} = 0, \quad (1.13)$$

$$\nabla \times \mathbf{H} = \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}}{\partial t} + \mathbf{J}, \quad (1.14)$$

and

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t}. \quad (1.15)$$

\mathbf{J} is the electric current density. Taking the curl of Equation (1.15) and the time derivative of Equation (1.14) we get

$$\nabla \times (\nabla \times \mathbf{E}) + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2} - \mu_0 \frac{\partial \mathbf{J}}{\partial t}. \quad (1.16)$$

We see that the left-hand side of this Equation is the simple wave Equation for a vacuum. The additional terms on the right-hand side are called *source terms*. The first source term involves polarisation charges and the second term involves conduction charges that are applicable to metallic materials. For a dielectric material $\mathbf{J} = 0$. Substituting the value for the polarisation obtained in Equation (1.10) into the wave Equation obtained in (1.16):

$$\nabla \times (\nabla \times \mathbf{E}) + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{Ne^2}{m} \left(\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \right) \frac{\partial^2 \mathbf{E}}{\partial t^2}. \quad (1.17)$$

Maxwell defined the electric displacement vector \mathbf{D} as

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$$

and for a medium with no localised charge density,

$$\nabla \cdot \mathbf{D} = 0.$$

For a spatially uniform medium,

$$\nabla \cdot \mathbf{P} = 0.$$

Therefore, $\nabla \cdot \mathbf{E} = 0$ and $\nabla \times (\nabla \times \mathbf{E}) = -\nabla^2 \mathbf{E}$. Equation (1.17) now becomes

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{Ne^2}{m} \left(\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \right) \frac{\partial^2 \mathbf{E}}{\partial t^2}.$$

Using the fact that $\mu_0 \epsilon_0 = \frac{1}{c^2}$,

$$\nabla^2 \mathbf{E} = \frac{1}{c^2} \left(1 + \frac{Ne^2}{m\epsilon_0} \left[\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \right] \right) \frac{\partial^2 \mathbf{E}}{\partial t^2}. \quad (1.18)$$

We see here that the wave Equation has a linear polarisation source term. If we consider a plane wave propagating in the z direction and expressed as

$$\mathbf{E} = \mathbf{E}_0 e^{i(kz - \omega t)}, \quad (1.19)$$

then

$$\nabla^2 \mathbf{E} = -k^2 \mathbf{E}$$

and

$$\frac{\partial^2 \mathbf{E}}{\partial t^2} = -\omega^2 \mathbf{E}.$$

From (1.18) we now get that

$$-k^2 \mathbf{E} = \frac{1}{c^2} \left(1 + \frac{Ne^2}{m} \left[\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \right] \right) (-\omega^2 \mathbf{E}),$$

and

$$k^2 = \frac{\omega^2}{c^2} \left[1 + \left(\frac{Ne^2}{m} \right) \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \right]. \quad (1.20)$$

From this we can conclude that a plane wave propagation in a homogeneous dielectric requires in general that the propagation constant k be a complex number. Defining the real and imaginary parts of k by

$$\tilde{k} = k_R + ik_I$$

and replacing this form into the expression for a harmonic wave as in Equation (1.19), we get

$$\mathbf{E} = \mathbf{E}_0 e^{i((k_R + ik_I z) - \omega t)} = \mathbf{E}_0 e^{k_I z} e^{i(k_R z - \omega t)}. \quad (1.21)$$

Taking the square of the magnitude of both sides gives

$$|\mathbf{E}|^2 = \mathbf{E}_0^2 e^{-2k_I z}.$$

The intensity of a field passing through a dielectric medium is given by [4]

$$I = \frac{1}{2} \epsilon_0 \frac{\eta}{v} |\mathbf{E}|^2, \quad (1.22)$$

where η is the refractive index of the medium, v is the speed of the wave in the medium and c is the speed of light. Therefore,

$$I = \frac{1}{2} \epsilon_0 \frac{\eta}{v} \mathbf{E}_0^2 e^{-2k_I z} = I_0 e^{-2k_I z} = I_0 e^{-\alpha z}. \quad (1.23)$$

where $\alpha = 2k_I$ is the absorption coefficient of the medium, and it determines how much of the incoming light is absorbed by the medium.

1.5 Linear Polarisation and Refractive Index

In this Section, we would like explain how the refractive index of light in a medium depends on the frequency of the light. Understanding this will help us later in Chapter 2 and 3 when we will discuss the effect of the different refractive indices for the fundamental and second-harmonic wave.

Since the propagation constant is complex (see Equation 1.20), so is the refractive index because

$$k = \frac{2\pi}{\lambda} = \frac{2\pi f}{v} = \left(\frac{\omega}{c} \right) \eta. \quad (1.24)$$

Writing out the real and imaginary parts of the refractive index, we get

$$\eta = \eta_R + i\eta_I. \quad (1.25)$$

So from (1.24)

$$k_R + ik_I = \left(\frac{\omega}{c}\right) (\eta_R + i\eta_I)$$

yielding the relations

$$k_R = \frac{\omega}{c} \eta_R,$$

and

$$k_I = \frac{\omega}{c} \eta_I.$$

From Equation (1.24) and (1.25), we can write

$$\eta^2 = (\eta_R + i\eta_I)^2 = \left(\frac{ck}{\omega}\right)^2,$$

then using Equation (1.20)

$$(\eta_R + i\eta_I)^2 = (\eta_R^2 - \eta_I^2) + i(2\eta_R\eta_I) = 1 + \left(\frac{Ne^2}{m\epsilon_0}\right) \frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}. \quad (1.26)$$

Re-writing the right-hand side in form of its real and imaginary parts by multiplying the numerator and denominator by the conjugate of the denominator, we get

$$(\eta_R^2 - \eta_I^2) + i(2\eta_R\eta_I) = 1 + \left(\frac{Ne^2}{m\epsilon_0}\right) \frac{\omega_0^2 - \omega^2 + i\omega\gamma}{(\omega_0^2 - \omega^2 - i\omega\gamma)(\omega_0^2 - \omega^2 + i\omega\gamma)}.$$

This can also be written as

$$(\eta_R^2 - \eta_I^2) + i(2\eta_R\eta_I) = 1 + \left(\frac{Ne^2}{m\epsilon_0}\right) \frac{\omega_0^2 - \omega^2 + i\omega\gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2}.$$

Separating the real and complex parts gives

$$(\eta_R^2 - \eta_I^2) + i(2\eta_R\eta_I) = 1 + \left(\frac{Ne^2}{m\epsilon_0}\right) \left[\frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} + \frac{i\omega\gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} \right]. \quad (1.27)$$

From Equation (1.27) we can see that

$$\eta_R^2 - \eta_I^2 = 1 + \left(\frac{Ne^2}{m\epsilon_0}\right) \left[\frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} \right] \quad (1.28)$$

and

$$2\eta_R\eta_I = \left(\frac{Ne^2}{m\epsilon_0}\right) \left[\frac{\gamma\omega}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} \right]. \quad (1.29)$$

The appearance of the mass in the denominator of these Equations shows that electronic oscillations are more important than ionic oscillations in determining the index of refraction, since electrons are much lighter than ions. A resonance frequency such as ω_0 for a dielectric medium, means that there is a high probability that an incident photon will be absorbed at that frequency ω_0 .

Equations (1.28) and (1.29) are used to solve for the optical coefficients η_R and η_I associated with a specific optical material. Plots of typical values of η_I and η_R versus frequency are shown in figure 1.1 and 1.2[2].

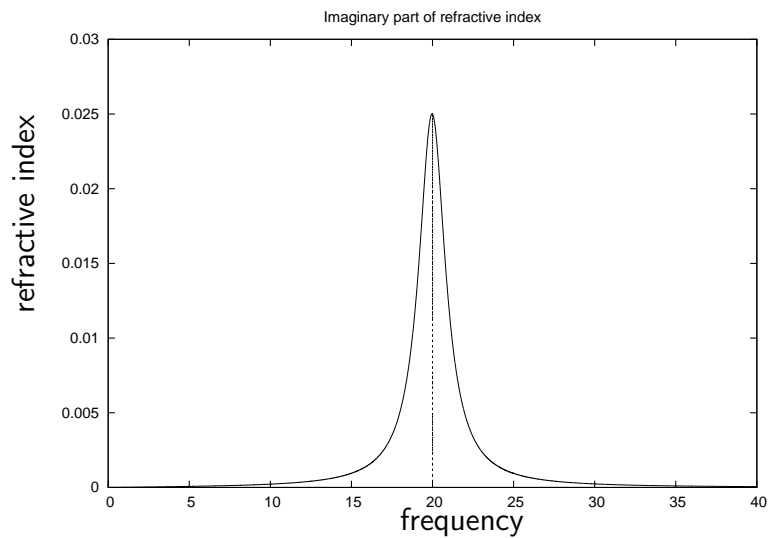


Figure 1.1: Imaginary part of the refractive index versus frequency, $\omega_0 = 20$

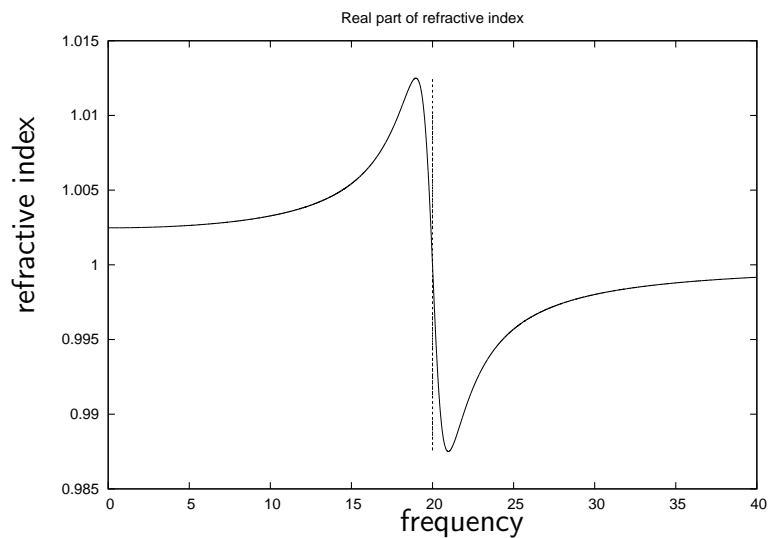


Figure 1.2: Real part of the refractive index versus frequency $\omega_0 = 20$

The value of η_I goes through a maximum at the resonance frequency ω_0 . Since the wavelength is approximately constant over the region where η_I has a significant value, it follows that the

absorption coefficient (see Equation (1.22)) also goes through a maximum at ω_0 . This frequency-sensitive absorption factor α suggests that the material absorption is largely dependent on the electronic characteristics of the material and how the bound electrons resonate within the material.

The plot of η_R versus ω in Figure 1.2 is referred to as the dispersion curve. It shows η_R gradually increasing for increasing ω , passing through a maximum, then decreasing rapidly to a minimum as it passes through ω_0 . It then increases again but returns to a lower value than that which it had before it approached the resonance, and again begins a gradual increase towards the next resonance. In any region where η_R increases with ω , or $\frac{d\eta_R}{d\omega} > 0$, the dispersion is referred to as normal dispersion. This is because $\frac{d\eta_R}{d\omega} > 0$ for most of the infrared, visible, and ultra spectral regions of most transparent materials. However, in some localised regions where there is a resonance, $\frac{d\eta_R}{d\omega} < 0$, we have a region of *anomalous* dispersion. In this case the absorption is very high and the electromagnetic radiation is rapidly extinguished[2].

A physical interpretation of the refractive index can be imagined as follows: the electric field of the incident wave polarises the atoms and sets the electrons in a forced sinusoidal motion. The acceleration of the charges radiates a new field that is superimposed on the initial one. The two fields vibrate at the same frequency, however, they are not in phase. When interfering they create, inside the material, a field that is not in phase with the incident field. It seems that the light propagates with a phase velocity that is not equal to c [5].

1.6 Relationship between Energy and Momentum of Photons

Since this Chapter explains the background of this essay, it will necessary for us to remind ourselves of the relationship between the energy and momentum of a photon. This will help us later in Section 2.2, when we will be discussing how the conservation of energy and momentum in the process of the generation of new frequency photons leads to different phase matching conditions.

The total energy E and magnitude of momentum for a particle with rest mass m is related by

$$E^2 = (mc^2)^2 + (pc)^2$$

since the photon is massless the energy-momentum relationship for a photon becomes

$$E^2 = (pc)^2 \implies E = pc,$$

The energy of an emitted photon is $E = \hbar\omega = hf = h\frac{c}{\lambda}$.

Consequently the magnitude of the momentum becomes

$$p = \hbar k = \frac{h}{\lambda} = h\frac{f}{c}.$$

Therefore, the energy and momentum of a photon depends only upon its frequency and consequently its wavelength. It is important for us to understand this relation now as it is relevant for our later discussion of energy and momentum conservation in nonlinear optical effects in Chapter 2.

1.7 Origin of Nonlinear Polarisation

We had earlier modelled the linear response of the electrons in a medium using the electron oscillation model in Section 1.3, similarly in this Section we will model the nonlinear atomic response of a noncentrosymmetric medium, (that is mediums lacking inversion symmetry) by allowing for nonlinearity in the restoring force exerted on the electrons. In this case, the restoring force on the electrons is not just

$$\mathbf{F} = -K\mathbf{r},$$

but

$$\mathbf{F} = -K_1\mathbf{r} - K_2\mathbf{r}^2 + \dots,$$

and we have neglected higher order terms. The Equation of motion of an electron then becomes[3]

$$\ddot{\mathbf{r}} + \gamma\dot{\mathbf{r}} + \omega_0^2\mathbf{r} + \mathbf{r} \cdot \tilde{a} \cdot \mathbf{r} = -e\mathbf{E}(t)/m, \quad (1.30)$$

where we have introduced a quadratic term, $\mathbf{r} \cdot \tilde{a} \cdot \mathbf{r}$, \tilde{a} being a third-rank tensor. For the sake of simplicity we will only consider only the one dimensional case so that Equation (1.30) can now be written as

$$\ddot{r} + \gamma\dot{r} + \omega_0^2r + ar^2 = -eE(t)/m. \quad (1.31)$$

We obtain this form by assuming that the restoring force is a nonlinear function of the displacement of the electron. Considering only the first and second term of the nonlinear expansion of the restoring force, we can understand the nature of the force by noting that it corresponds to a potential energy function of the form

$$U = - \int F dr = \frac{1}{2}m\omega_0^2r^2 + \frac{1}{3}mar^3. \quad (1.32)$$

Here the first term corresponds to a harmonic potential and the second term corresponds to an anharmonic correction term. This model corresponds to the physical situation of electrons in real materials, because the actual potential well that the atomic electron feels is not perfectly parabolic[3]. In this case, let us assume that the applied optical field is of the form

$$E(t) = E_1e^{i\omega_1t} + E_2e^{i\omega_2t} + c.c..$$

No general solution to Equation (1.31) exist analytically. However, if the applied field is sufficiently weak, the nonlinear term ar^2 will be much smaller than the linear term ω_0^2r for any displacement r . In order to solve Equation (1.31) we use the idea of perturbation expansion [3]. This means we now replace $E(t)$ by $\lambda E(t)$, where λ is a parameter that ranges continuously between zero and one, and at the end of the calculation will be set equal to one. The expansion parameter λ thus characterises the strength of the perturbation. Equation (1.31) then becomes

$$\ddot{r} + \gamma\dot{r} + \omega_0^2r + ar^2 = -\lambda eE(t)/m. \quad (1.33)$$

Note here that λ in this Section has nothing to with wavelength, and we just employ this method because we need to solve the Equation (1.31).

We now seek a solution to Equation (1.33) in the form of a power series expansion, that is

$$r = \lambda r^{(1)} + \lambda^2 r^{(2)} + \lambda^3 r^{(3)} + \dots \quad (1.34)$$

Using only the first and second term, substituting into Equation (1.33), and equating the coefficients of the first and second powers of λ , we obtain the following Equations for the coefficients of λ^1 and λ^2 respectively as

$$\ddot{r}^{(1)} + \gamma \dot{r}^{(1)} + \omega_0^2 r^{(1)} = -eE(t)/m, \quad (1.35)$$

$$\ddot{r}^{(2)} + \gamma \dot{r}^{(2)} + \omega_0^2 r^{(2)} + a [r^{(1)}]^2 = 0. \quad (1.36)$$

Notice that Equation (1.35) is the same as the Equation of motion describing the linear response of the atoms to the electric field we obtained in Equation (1.6), and we have already found the solution for this in Equation (1.7). In order to solve Equation (1.36), we choose a solution of the form

$$r^{(1)}(t) = r^{(1)}(\omega_1)e^{i\omega_1 t} + r^{(1)}(\omega_2)e^{i\omega_2 t} + c.c., \quad (1.37)$$

where the amplitudes $r^{(1)}(\omega_j)$ have the form (see Equation (1.7))

$$r^{(1)}(\omega_j) = -\frac{e}{m} \frac{E_j}{D(\omega_j)}, \quad (1.38)$$

and we have introduced the complex denominator function

$$D(\omega) = \omega_0^2 - \omega^2 - i\omega\gamma. \quad (1.39)$$

Squaring Equation (1.37) gives

$$\begin{aligned} [r^{(1)}(t)]^2 &= [r^{(1)}(\omega_1)]^2 e^{-2i\omega_1 t} + 2r^{(1)}(\omega_1)r^{(1)}(\omega_2)e^{-i(\omega_2+\omega_1)t} + [r^{(1)}(\omega_2)]^2 e^{-2i\omega_2 t} \\ &+ 2r^{(1)}(\omega_1)r^{(1)*}(\omega_2)e^{-i(\omega_1-\omega_2)t} + 2r^{(1)*}(\omega_1)r^{(1)}(\omega_2)e^{-i(\omega_2-\omega_1)t} \\ &+ 2r^{(1)*}(\omega_1)r^{(1)*}(\omega_2)e^{-i(-\omega_1-\omega_2)t} + [r^{(1)*}(\omega_1)]^2 e^{-i(-2\omega_1)t} \\ &+ 2r^{(1)}(\omega_1)r^{(1)*}(\omega_1) + 2r^{(1)}(\omega_2)r^{(1)*}(\omega_2). \end{aligned}$$

Notice that the square of $r^{(1)}(t)$ contains the frequencies $\pm 2\omega_1$, $\pm 2\omega_2$, $\pm(\omega_1 + \omega_2)$, $\pm(\omega_1 - \omega_2)$ and 0. We see here, and for the first time, the source of the second-harmonic generation. Let us briefly explain this, when we considered the second order term in the nonlinear displacement of the electrons by the applied force, and solved the Equation of motion for the electrons, we amongst other frequencies, observed the generation of a frequency doubling that of the frequency of the incident light ($2\omega_1, 2\omega_2$). So, the generation of the second-harmonic frequency is source from the the nonlinear displacement of the electrons and consequently from the susceptibility of the material.

In order to determine the response of the material at frequency $2\omega_1$, for instance, we must solve the Equation

$$\ddot{r}^{(2)} + \gamma \dot{r}^{(2)} + \omega_0^2 r^{(2)} = -a [r^{(1)}]^2, \quad (1.40)$$

and this implies solving

$$\ddot{r}^{(2)} + \gamma \dot{r}^{(2)} + \omega_0^2 r^{(2)} = -a \left[\frac{e^2}{m^2} \frac{E_1^2 e^{-2i\omega_1 t}}{D^2(\omega_1)} \right]. \quad (1.41)$$

We seek a solution of the form

$$r^{(2)}(t) = r^{(2)}(2\omega_1) e^{-2\omega_1 t}. \quad (1.42)$$

Substituting Equation (1.42) into Equation (1.41) we get that

$$(-4\omega_1^2 - 2i\gamma\omega_1 + \omega_0^2) r^{(2)}(2\omega_1) e^{-2\omega_1 t} = -a \left[\frac{e^2}{m^2} \frac{E_1^2 e^{-2i\omega t}}{D^2(\omega_1)} \right] \quad (1.43)$$

and this implies

$$r^{(2)}(2\omega_1) = -a \left[\frac{e^2}{m^2} \frac{E_1^2}{D(2\omega_1)D^2(\omega_1)} \right] \quad (1.44)$$

where

$$D(2\omega_1) = -4\omega_1^2 - 2i\omega_1\gamma + \omega_0^2. \quad (1.45)$$

Analogously, the amplitudes of the response at frequency $2\omega_2$ is found to be

$$r^{(2)}(2\omega_2) = -a \left[\frac{e^2}{m^2} \frac{E_1^2}{D(2\omega_2)D^2(\omega_1)} \right]. \quad (1.46)$$

The linear contribution to polarisation from Equation (1.1) is given in a more general form by

$$P^{(1)}(\omega_1) = -Ner^{(1)}(\omega_j)$$

then using Equation (1.38), we can now write $P^{(1)}(\omega_1)$ as

$$P^{(1)}(\omega_1) = \frac{Ne^2 E_j}{mD(\omega_j)}, \quad (1.47)$$

From Equation (1.47), we define the linear susceptibility in general as

$$\chi^{(1)}(\omega_i) = \frac{Ne^2}{mD(\omega_j)}.$$

The linear polarisation in a more general form is defined by

$$P^{(1)}(\omega_1) = \chi^{(1)}(\omega_j) E(\omega_i). \quad (1.48)$$

The nonlinear polarisation and susceptibilities are calculated in an analogous manner. The nonlinear susceptibility describing second-harmonic generation $\chi^{(2)}$ is defined by the relation

$$P^{(2)}(2\omega_1) = \chi^{(2)}(2\omega_1, \omega_1, \omega_1) E(\omega_1)^2. \quad (1.49)$$

Here ,

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{N(e^3/m^2)a}{D(2\omega_1)D^2(\omega_1)} \quad (1.50)$$

and $P^{(2)}(2\omega_1)$ is the nonlinear polarisation source for the second-harmonic generation.

From Equation (1.49), we see that the nonlinear polarisation source for the second-harmonic generation depends on the square of the incident electric field. In order to maximise this, the applied electric field must have a high intensity. That is why the invention of lasers with high intensity light brought to the fore, the nonlinear optical effects in materials. Also, the nonlinear polarisation source for the second-harmonic generation depends on the nonlinear susceptibility $\chi^{(2)}(2\omega_1, \omega_1, \omega_1)$ expressed in Equation (1.50), in the denominator of this susceptibility, we see from Equation (1.39) and (1.45), that the second order nonlinear susceptibility ($\chi^{(2)}(2\omega_1, \omega_1, \omega_1)$) may be resonantly enhanced at the frequencies ($\omega_0 \approx \omega_1$ and $\omega_0 \approx 2\omega_1$). In resonant enhancement processes the frequency of the lasers is tuned, so that it is close to the resonant frequency of the material. This helps to increase the susceptibility.

This Section helps to understand that if we consider the application of an intense electric field to a medium, the expression for a linear polarisation obtained in Equation (1.11) does not hold. From the electron model we would say that the displacement r of the electrons is no longer small since the field is intense and so we must consider at least the quadratic term in the nonlinear expansion of the elastic restoring force. Doing this, we discovered the polarisation of a nonlinear medium can even be expressed as a power series in the applied electric field and this leads us to Chapter 2 where we will look at the different second order nonlinear optical effects.

2. Second Order Nonlinear Optical Phenomena

2.1 Nonlinear Polarisation

The linear polarisation \mathbf{P} of a material depends upon the strength of the applied field as

$$\mathbf{P} = \epsilon_0 \chi^{(1)} \mathbf{E}. \quad (2.1)$$

Where $\chi^{(1)}$ is the linear electric susceptibility, a useful parameter when considering the response of an material to applied electric field. In nonlinear optics, where the electric field strength is high enough, Equation (2.1) can then be generalised by expressing it as a power series

$$P = \epsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots). \quad (2.2)$$

The polarisation effect makes a significant contribution to the nonlinear optical process since it results from charges oscillating within the medium due to the incident oscillating electric field and re-radiating, thereby adding to that field. For simplicity, we have expressed P and E as scalar quantities. Also since Equation (2.2) is time dependent, we have assumed that the response of the material is instantaneous to the applied field. For this assumption to be true, the medium must have no loss or dispersion[2].

2.1.1 Nonlinear Wave Equation

We had earlier derived the wave equation of a dielectric medium in Equation 1.15, we now use Maxwell's Equations to derive the nonlinear wave Equation, this Equation shows that the nonlinear polarisation source is frequency dependent. We consider the time-spatial variation of an optical field according to Maxwell's Equation

$$\nabla \times \mathbf{H} = \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}}{\partial t} \quad (2.3)$$

and

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t}. \quad (2.4)$$

In general, \mathbf{E} , \mathbf{P} and \mathbf{H} may be arbitrary time-spatial functions that can be expressed in Fourier integral form [6]:

$$\mathbf{E}(t, r) = \int_{-\infty}^{\infty} \mathbf{E}(\omega, r) e^{-i\omega t} d\omega,$$

$$\mathbf{H}(t, r) = \int_{-\infty}^{\infty} \mathbf{H}(\omega, r) e^{-i\omega t} d\omega,$$

and

$$\mathbf{P}(t, r) = \int_{-\infty}^{\infty} \mathbf{P}(\omega, r) e^{-i\omega t} d\omega.$$

Here, r represents the spatial variables. The physical meaning of these expressions is that any given optical field can always be viewed as a certain superposition of infinite monochromatic Fourier components[6].

Now, for a given function $f(t)$, the Fourier transform of its derivative $f'(t)$ is given by

$$\begin{aligned} F(f'(t)) &= \int_{-\infty}^{\infty} f'(t) e^{i\omega t} dt \\ &= e^{i\omega t} f(t) - i\omega \int_{-\infty}^{\infty} e^{i\omega t} f(t) dt \\ &= -i\omega f(\omega). \end{aligned} \quad (2.5)$$

Here $f(t)$ tends to zero at $\pm \infty$ (as it must since $\int_{-\infty}^{\infty} |f(t)| dt$ must be finite [7]). So taking the derivative of the Fourier transforms in Equation (2.1.1), we get that

$$\int_{-\infty}^{\infty} \frac{\partial \mathbf{E}(t, r)}{\partial t} e^{i\omega t} dt = -i\omega \mathbf{E}(\omega, r),$$

$$\int_{-\infty}^{\infty} \frac{\partial \mathbf{H}(t, r)}{\partial t} e^{i\omega t} dt = -i\omega \mathbf{H}(\omega, r),$$

and

$$\int_{-\infty}^{\infty} \frac{\partial \mathbf{P}(t, r)}{\partial t} e^{i\omega t} dt = -i\omega \mathbf{P}(\omega, r). \quad (2.6)$$

With these transformations Equation (2.3) becomes

$$\nabla \times \mathbf{H}(\omega, r) = -\epsilon_0 i\omega \mathbf{E}(\omega, r) - i\omega \mathbf{P}^L(\omega, r) - i\omega \mathbf{P}^{NL}, \quad (2.7)$$

where we have expressed the polarisation \mathbf{P} in terms of its linear term \mathbf{P}^L and nonlinear term \mathbf{P}^{NL} . We have also assumed that

$$\mathbf{P}(\omega, r) = \mathbf{P}^L(\omega, r) + \mathbf{P}^{NL}(\omega, r).$$

Similarly Equation (2.4) becomes

$$\nabla \times \mathbf{E}(\omega, r) = \mu_0 i\omega \mathbf{H}(\omega, r). \quad (2.8)$$

Taking the curl of Equation (2.8) and using (2.7), we get

$$\nabla \times (\nabla \times \mathbf{E}(\omega, r)) = \mu_0 \epsilon_0 \omega^2 \mathbf{E}(\omega, r) + \mu_0 \omega^2 [\mathbf{P}^L(\omega, r) + \mathbf{P}^{NL}(\omega, r)]. \quad (2.9)$$

Considering that

$$\mathbf{P}^L(\omega, r) = \epsilon_0 \chi^{(1)}(\omega) \mathbf{E}(\omega, r) \quad (2.10)$$

and

$$\epsilon(\omega) = \epsilon_0 [1 + \chi^{(1)}(\omega)], \quad (2.11)$$

where $\epsilon(\omega)$ is the linear dielectric constant at frequency ω , Equation (2.9) can then be written as

$$\nabla \times (\nabla \times \mathbf{E}(\omega, r)) - \mu_0 \epsilon_0 \omega^2 \epsilon(\omega) \mathbf{E}(\omega, r) = \mu_0 \omega^2 \mathbf{P}^{NL}(\omega, r). \quad (2.12)$$

Equation (2.12) is the fundamental Equation for $\mathbf{E}(\omega, r)$ and $\mathbf{P}^{NL}(\omega, r)$ and is called the nonlinear wave Equation. So for a weak incident optical field $\mathbf{P}^{NL}(\omega, r) \approx 0$, Equation (2.12) becomes the linear wave Equation

$$\nabla \times (\nabla \times \mathbf{E}(\omega, r)) - \mu_0 \epsilon_0 \omega^2 \epsilon(\omega) \mathbf{E}(\omega, r) = 0. \quad (2.13)$$

In this case, the polarisation response of a material to a given monochromatic component $\mathbf{E}(\omega, r)$ of the applied field is only represented by the dielectric constant $\epsilon(\omega)$; the other frequency components of the field have no influence on either $\mathbf{P}^L(\omega, r)$ or $\mathbf{E}(\omega, r)$. Whereas, if the applied field is an intense laser field, the second and/or third order components expressed by Equation (2.2) may no longer be neglected. Then Equation (2.12) becomes a nonlinear differential Equation, and the term $\mathbf{P}^{NL}(\omega, r)$ can be recognised as a source that can emit coherent radiation at a new frequency[6].

2.2 Second Order Nonlinear Optical Processes

2.2.1 Second-Harmonic Generation

We now know that with an intense laser field, the polarisation of the material is a nonlinear function of the electric field, so what happens in second-harmonic generation is that, an intense laser beam of angular frequency ω is passed through a crystal with nonzero value of $\chi^{(2)}$, such that the beam emerging from the crystal contains the the angular frequencies ω of the input beam and also 2ω , twice the frequency of the input beam. This can be shown to occur by considering the second order nonlinear polarisation term (2.2). Let us consider the case in which two input photons have different frequencies ω_1 and ω_2 , where $\omega_1 > \omega_2$.

$$E = E_1 e^{-i\omega_1 t} + E_1^* e^{i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_2^* e^{i\omega_2 t}. \quad (2.14)$$

We consider the second order nonlinear polarizability as

$$\begin{aligned} P^{(2)} &= \epsilon_0 \chi^{(2)} E^2 \\ &= \epsilon_0 \chi^{(2)} [E_1^2 e^{-i(2\omega_1)t} + (E_1^*)^2 e^{i(2\omega_1)t} + E_2^2 e^{-i(2\omega_2)t} + (E_2^*)^2 e^{i(2\omega_2)t} \\ &\quad + 2E_1 E_1^* + 2E_2 E_2^* + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} + 2E_1^* E_2^* e^{i(\omega_1 + \omega_2)t} \\ &\quad + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + 2E_1^* E_2 e^{i(\omega_1 - \omega_2)t}]. \end{aligned}$$

The result includes the second-harmonic terms with $2\omega_1$ and $2\omega_2$. The direct current terms represents a time independent factor that produces no oscillating electromagnetic radiation. The sum-frequency generation is the addition of the two fundamental frequencies $\omega_1 + \omega_2$ and the difference frequency generation $\omega_1 - \omega_2$.

Let us look at this different optical frequency mixing effects from a quantum theory of radiation stand point. The advantage of considering the quantum theory of radiation is that it not only describes the wave property of the optical field but also involves transfer of energy and momentum.

Before we go into describing how the second order nonlinear optical effects can be described from the quantum mechanical stand point, it is pertinent to recall how the quantum theory of radiation works.

An interaction between an optical field and a medium is always accompanied by

1. a quantum transition of molecules of the medium among their different states.
2. a change of photon distribution. In order to properly describe this process we must introduce the key concept of the so-called *intermediate state*.

In this intermediate state of the whole system, the degeneracy among the different modes is changed while the molecules have left their original state which is usually the ground state. At this point we cannot say exactly which excited state a molecule is, but we can distribute the state of a molecule to all possible excited states with a certain distribution probability. In this case, it is convenient to introduce a *virtual state* in the energy level diagram to represent this intermediate state. Based on the concept of intermediate state, as well as a representation of a virtual energy level, we can now explain the second order nonlinear optical effects.

In the case of the second-harmonic-generation, two photons of frequency ω leaves the ground state to an intermediate, while returning to its initial state there is an emission of a second-harmonic photon. As shown in the diagram below [6].

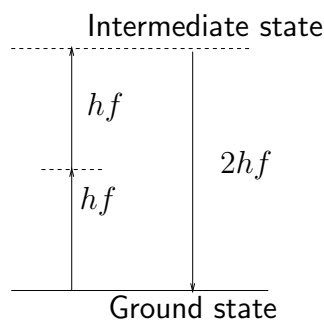


Figure 2.1: Energy level description of second-harmonic generation from a quantum standpoint.

In Figure 2.1, h is the Planck constant and f is the frequency of the photon.

Since the transit time of the molecules in the intermediate state is extremely short about $10^{-16}s$, the above processes occur simultaneously and instantaneously[3]. One can find that at the beginning and end of this process, the quantum states of this processes are unchanged (the

energy and momentum of the molecules are unchanged) therefore the conservation of energy and momentum should hold among the annihilated photons and created photon(s).

Assuming the frequencies of the fundamental and second-harmonic waves are f_1 and f_2 respectively, then conservation of energy and momentum gives us that

$$hf_2 = 2hf_1, \quad (2.15)$$

and

$$\mathbf{k}_2 = \mathbf{k}_1 + \mathbf{k}_1. \quad (2.16)$$

Here \mathbf{k}_1 and \mathbf{k}_2 are the wave vectors of these two waves and h is Planck's constant. For collinear propagation of the two waves, Equation (2.16) gives that

$$\begin{aligned} \frac{2\pi}{\lambda_2} &= 2 \cdot \frac{2\pi}{\lambda_1} \\ \implies \frac{2\pi f_2}{v_2} &= 2 \cdot \frac{2\pi f_1}{v_1} \end{aligned} \quad (2.17)$$

where v is the corresponding speed of the waves, and this implies that

$$\frac{\omega_2}{c} \eta(f_2) = 2 \frac{\omega_1}{c} \eta(f_1), \quad (2.18)$$

where $\eta(f_1)$ and $\eta(f_2)$ are the refractive indices of the medium at the frequencies f_1 and f_2 respectively.

From Equation (2.15), we know that $2\omega_1 = \omega_2$, hence Equation (2.18) leads to the so called phase matching condition

$$\eta(2f_1) = \eta(f_1). \quad (2.19)$$

So we see that the phase-matching condition which we will further explain in Section 3.2, is a natural result of the conservation of energy and momentum from the stand-point of quantum theory of radiation.

2.2.2 Optical Sum Frequency Generation

Within the same framework we will look at the sum-frequency generation from the quantum mechanical point of view. In this case the elementary process is composed of two steps, in the first step, two photons one from ω_1 and the other from ω_2 is simultaneously annihilated as a molecule leaves its ground state to an intermediate state. On returning, one photon is created and added to the ω_3 field. The energy level diagram for sum-frequency generation is shown in Figure 2.2

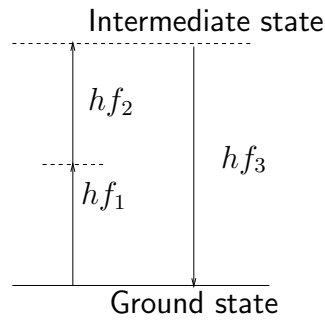


Figure 2.2: Energy level description of sum-frequency generation from a quantum standpoint.

There should be conservation of energy and momentum at the beginning and end of the process, so we then have

$$hf_3 = hf_1 + hf_2, \quad (2.20)$$

and

$$\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2, \quad (2.21)$$

where \mathbf{k}_1 , \mathbf{k}_2 and \mathbf{k}_3 are the wave vectors of the these three waves. From Equation (2.21), we see that the phase matching condition requires that

$$\frac{\omega_3}{c}\eta(f_3) = \frac{\omega_1}{c}\eta(f_1) + \frac{\omega_2}{c}\eta(f_2), \quad (2.22)$$

and this implies that

$$\omega_3\eta(f_3) = \omega_1\eta(f_1) + \omega_2\eta(f_2).$$

2.2.3 Optical Difference-Frequency Generation

Using the same assumption that the incident optical field contains two frequency components ω_1 and ω_2 and $\omega_1 > \omega_2$ a new radiation wave at the frequency $\omega_3 = \omega_1 - \omega_2$ can be generated and is explained from the quantum point that, there is an annihilation of one high frequency photon as a photon leaves its ground state to an intermediate state and simultaneously there is the creation of two lower frequencies (hf_2 and hf_3) photons when the molecule is returning to its ground state.

In a similar way, as the other two frequency conversions, there must be conservation of energy and momentum in these processes so,

$$hf_1 = hf_2 + hf_3,$$

and

$$\mathbf{k}_1 = \mathbf{k}_2 + \mathbf{k}_3.$$

These Equations leads to the phase matching condition of

$$\frac{\omega_1}{c}\eta(f_1) = \frac{\omega_2}{c}\eta(f_2) + \frac{\omega_3}{c}\eta(f_3).$$

which implies that

$$\omega_1\eta(f_1) = \omega_2\eta(f_2) + \omega_3\eta(f_3).$$

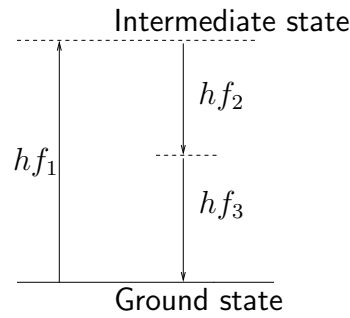


Figure 2.3: Energy level description of difference-frequency generation from a quantum standpoint.

2.2.4 Nonlinear Crystals for Second-harmonic generation

There are some basic requirements for nonlinear crystals to support second-harmonic generation. We will briefly discuss some of these,

1. **Noncentrosymmetry:** In the electric dipole approximation for nonlinear media described in Section 1.7, isotropic media and centrosymmetric crystals cannot be used to generate second order nonlinear effects. Therefore, the media for second-harmonic generation should be the crystals lacking symmetry inversion such that $U(-x) = U(x)$ and the potential energy is of the form so, for centrosymmetric media the potential energy should be of the form

$$U = \frac{1}{2}m\omega_0^2x^2 - \frac{1}{4}mbx^4 + \dots \quad (2.23)$$

So we see that for a centrosymmetric media the term ar^2 in Equation (1.31) is replaced by a nonlinear term br^3 and the second order nonlinear susceptibility vanishes.

2. **Phase-matching:** We have proved theoretically that only when the phase matching condition of $\eta(2f_1) = \eta(f_1)$ is achieved can the second-harmonic generation be efficiently obtained. For a normal dispersion (see Section 1.5), $\eta(2f_1)$ is greater than $\eta(f_1)$ and we have to choose a special direction within a given crystal so that the dispersion effect of refractive index is compensated by the birefringence effect(see Section 3.2.1). The angle between this special direction and the optical axis of the crystal is termed the phase matching angle. phase matching angle is explained further in Section 3.2.2.
3. **Larger effective second-order susceptibility value:** Since the efficiency of the second-harmonic generation is proportional to the square of the effective second order susceptibility value $\chi^{(2)}$, so crystals with a larger value of $\chi^{(2)}$ are preferred. This dependence is seen from Equation (3.15) and (3.30).
4. **High optical damage threshold:** To ensure a higher and incident optical power, all crystals for second-harmonic generation should possess the best optical quality and high transmittivity for both the fundamental and second-harmonic waves.

3. Second-Harmonic Generation

In this Chapter we try to solve using Maxwell's Equation for the intensity of the second-harmonic wave and explain the ideal ways of obtaining an efficient second-harmonic generation.

3.1 Intensity of a Second-Harmonic Wave

From Maxwell's Equations, we see that the wave Equation (see Equation (1.17)) for a non conducting medium is

$$\nabla^2 E - \epsilon_0 \mu_0 \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}. \quad (3.1)$$

We can then deduce that the second-harmonic term in the polarisation P should generate a second-harmonic contribution to the electric field E . This means that an electric field of frequency ω in a non-centrosymmetric medium, will through the nonlinear polarisation $P^{NL}(2\omega)$ generate an electric field at the second-harmonic frequency 2ω . Let us try to derive the intensity of the electric field at the second-harmonic. We write the second-harmonic field as

$$E = \frac{1}{2} [E_{2\omega}(z)e^{-i(2\omega t - k_{2\omega}z)} + E_{2\omega}^*(z)e^{i(2\omega t - k_{2\omega}z)}], \quad (3.2)$$

where $k_{2\omega} = \frac{2\omega}{c}\eta(2\omega)$ and $\eta(2\omega) = (\epsilon_{2\omega}/\epsilon_0)^{1/2}$ is the refractive index of the medium for radiation of frequency 2ω . The wave Equation for the second-harmonic generation can then be derived. First the first term in the left hand side of Equation (3.1) becomes

$$\nabla^2 E_{2\omega} = \frac{1}{2} \left[\left(\frac{d^2 E_{2\omega}}{dz^2} + ik_{2\omega} \frac{dE_{2\omega}}{dz} \right) + ik_{2\omega} \left(\frac{dE_{2\omega}}{dz} - k_{2\omega}^2 E_{2\omega} \right) \right] e^{-i(2\omega t - k_{2\omega}z)} + c.c.. \quad (3.3)$$

Assuming the amplitude $E_{2\omega}$ to be slowly varying in z we make an approximation by neglecting second derivatives of $E_{2\omega}$, to get

$$\nabla^2 E_{2\omega} = \frac{1}{2} \left(2ik_{2\omega} \frac{dE_{2\omega}}{dz} - k_{2\omega}^2 E_{2\omega} + c.c. \right), \quad (3.4)$$

which together with

$$\epsilon_0 \mu_0 \frac{\partial^2 E_{2\omega}}{\partial t^2} = -2\epsilon_0 \mu_0 \omega^2 [E_{2\omega} e^{-i(2\omega t - k_{2\omega}z)} + c.c.], \quad (3.5)$$

gives the left side of Equation (3.1) as

$$\nabla^2 E_{2\omega} - \epsilon_0 \mu_0 \frac{\partial^2 E_{2\omega}}{\partial t^2} \approx \left(ik_{2\omega} \frac{dE_{2\omega}}{dz} - \frac{1}{2} (k_{2\omega}^2 - 4\epsilon_0 \mu_0 \omega^2) E_{2\omega} \right) e^{-i(2\omega t - k_{2\omega}z)} + c.c. \quad (3.6)$$

For the second-harmonic field Equation in (3.2), the right side of Equation (3.1) has both linear (L) and nonlinear (NL) contributions at frequency 2ω :

$$P = \frac{1}{2} [P_{2\omega}^{(L)}(z)e^{i(2\omega t - k_{2\omega}z)} + P_{2\omega}^{(L)*}(z)e^{i(2\omega t - k_{2\omega}z)} + P_{2\omega}^{(NL)}(z)e^{2i(\omega t - k_{\omega}z)} + P_{2\omega}^{(NL)*}(z)e^{2i(\omega t - k_{\omega}z)}] \quad (3.7)$$

and

$$\mu_0 \frac{\partial^2 P}{\partial t^2} = -2\mu_0\omega^2 [P_{2\omega}^{(L)}(z)e^{i(2\omega t - k_{2\omega}z)} + P_{2\omega}^{(L)*}(z)e^{i(2\omega t - k_{2\omega}z)} - 2\mu_0\omega^2 [P_{2\omega}^{(NL)}(z)e^{2i(\omega t - k_{\omega}z)} + P_{2\omega}^{(NL)*}(z)e^{2i(\omega t - k_{\omega}z)}]. \quad (3.8)$$

Combining the real parts of Equations (3.6) and (3.8), we get

$$\left(ik_{2\omega} \frac{dE_{2\omega}}{dz} - \frac{1}{2} (k_{2\omega}^2 - 4\epsilon_0\mu_0\omega^2) E_{2\omega} \right) e^{-i(2\omega t - k_{2\omega}z)} = -2\mu_0\omega^2 P_{2\omega}^L(z)e^{i(2\omega t - k_{2\omega}z)} - 2\mu_0\omega^2 P_{2\omega}^{NL}(z)e^{2i(\omega t - k_{\omega}z)}. \quad (3.9)$$

We know that $P = \epsilon_0\chi E$, which at a frequency of 2ω becomes

$$P_{2\omega}^{(L)} = \epsilon_0\chi(2\omega)E_{2\omega} \quad (3.10)$$

and

$$k_{2\omega}^2 = (2\omega)^2\epsilon_{2\omega}\mu_0 = 4\omega^2\epsilon_0\mu_0\eta^2(2\omega) = 4\omega^2\epsilon_0\mu_0 [1 + \chi(2\omega)], \text{ see Equation (2.11)} \quad (3.11)$$

We have also used the relation; speed of light in vacuum $c = \frac{1}{\sqrt{\mu_0\epsilon_0}}$ and the speed of light in a medium at frequency 2ω is $v_{2\omega} = \frac{1}{\sqrt{\epsilon_{2\omega}\mu_0}}$.

We have taken $\chi_{2\omega}$ to be real because we are assuming absorption and other loss processes to be negligible. The use of these relations in Equations (3.9) results in the Equation

$$ik_{2\omega} \frac{dE_{2\omega}}{dz} e^{i(2\omega t - k_{2\omega}z)} = -2\mu_0\omega^2 P_{2\omega}^{NL}(z)e^{2i(\omega t - k_{\omega}z)}, \quad (3.12)$$

or

$$\begin{aligned} \frac{dE_{2\omega}}{dz} &= \frac{2i\mu_0\omega^2}{k_{2\omega}} P_{2\omega}^{(NL)} e^{i(2\omega t - k_{2\omega}z - 2\omega t + 2k_{\omega}z)} \\ &= i\omega \sqrt{\mu_0/\epsilon_{2\omega}} P_{2\omega}^{(NL)} e^{i(2k_{\omega} - k_{2\omega})z}, \end{aligned} \quad (3.13)$$

and this relates the second-harmonic field to the nonlinear polarisation. Finally it is convenient to define a quantity

$$P_{2\omega}^{(NL)} = dE_{2\omega}^2(z), \quad (3.14)$$

where d represents (see Equation (1.49))

$$d = \epsilon_0 \chi^{(2)}(2\omega, \omega, \omega). \quad (3.15)$$

Thus we can write Equation (3.13) as

$$\frac{dE_{2\omega}}{dz} = i\omega \sqrt{\frac{\mu_0}{\epsilon_{2\omega}}} dE_{\omega}^2(z) e^{i\Delta kz}, \quad (3.16)$$

where

$$\Delta k = 2k_{\omega} - k_{2\omega} = 2\omega \sqrt{\epsilon_0 \mu_0} [\eta(\omega) - \eta(2\omega)]. \quad (3.17)$$

The solution to Equation (3.16) gives the second-harmonic field amplitude $E_{2\omega}(z)$. In order to solve the Equation, we need to know $E_{\omega}(z)$. To simplify our work, we make the approximation that there is little attenuation of the fundamental wave so that E_{ω} is a constant

$$E_{\omega}(z) \approx E_{\omega}(0). \quad (3.18)$$

This approximation is often called *small signal limit*. Within this approximation, the signal of the second-harmonic wave generated is still small. In practice as the conversion from a fundamental to a second-harmonic wave continues in the medium, there is depletion of the fundamental wave and this approximation is no longer a good one.

With this approximation (3.16) can be integrated to obtain

$$E_{2\omega}(z) \approx i\omega \sqrt{\frac{\mu_0}{\epsilon_{2\omega}}} dE_{\omega}^2(0) \int_0^z e^{i\Delta kz} dz \quad (3.19)$$

$$= i\omega \sqrt{\frac{\mu_0}{\epsilon_{2\omega}}} dE_{\omega}^2(0) \left(\frac{1}{i\Delta k} (e^{i\Delta kz} - 1) \right). \quad (3.20)$$

Now $\frac{1}{i\Delta k} (e^{i\Delta kz} - 1)$ can be written as

$$\frac{1}{i\Delta k} (e^{i\Delta kz} - 1) = \frac{e^{i\Delta kz/2}}{i\Delta k} (e^{i\Delta kz/2} - e^{-i\Delta kz/2}). \quad (3.21)$$

Equation (3.21) becomes

$$\frac{1}{i\Delta k} (e^{i\Delta kz} - 1) = 2e^{i\Delta kz/2} \frac{1}{\Delta k} \sin \frac{1}{2} \Delta kz \quad (3.22)$$

$$= ze^{i\Delta kz/2} \left(\frac{\sin \frac{1}{2} \Delta kz}{\frac{1}{2} \Delta kz} \right). \quad (3.23)$$

So with the approximation that the fundamental wave is unattenuated we have

$$E_{2\omega}(z) = i\omega \sqrt{\frac{\mu_0}{\epsilon_{2\omega}}} dE_{\omega}^2(0) z e^{i\Delta kz/2} \left(\frac{\sin \frac{1}{2} \Delta kz}{\frac{1}{2} \Delta kz} \right). \quad (3.24)$$

Equation (3.24) describes the amplitude of the second-harmonic wave, under the approximation that the applied field is not attenuated inside the medium.

Suppose the nonlinear crystal is of length L as in Figure (2.3) . The second-harmonic field at the exit face of the crystal is then given by Equation (3.23) with $z = L$. Thus Equation (3.24) implies that

$$|E_{2\omega}(L)|^2 = \frac{\mu_0\omega^2}{\epsilon_{2\omega}}|E_\omega(0)|^4 L^2 \left(\frac{(\sin\frac{1}{2})\Delta kL}{\frac{1}{2}\Delta kL} \right). \quad (3.25)$$

The intensities of the fields at ω and 2ω as given by Equation (1.22) are

$$I_\omega = \frac{1}{2}\sqrt{\frac{\epsilon_\omega}{\mu_0}}|E_\omega|^2, \quad (3.26)$$

$$I_{2\omega} = \frac{1}{2}\sqrt{\frac{\epsilon_{2\omega}}{\mu_0}}|E_{2\omega}|^2. \quad (3.27)$$

It follows from (3.25) that

$$\begin{aligned} I_{2\omega}(L) &= \frac{2\mu_0^{3/2}d^2\omega^2}{\epsilon_\omega\sqrt{\epsilon_{2\omega}}}|E_\omega(0)|^4 \frac{1}{4}\epsilon_\omega/\mu_0 L^2 \left(\frac{\sin\frac{1}{2}\Delta kL}{\frac{1}{2}\Delta kL} \right)^2 \\ &= \frac{2\mu_0^{3/2}d^2\omega^2}{\epsilon_\omega\sqrt{\epsilon_{2\omega}}}I_\omega^2(0)L^2 \left(\frac{\sin\frac{1}{2}\Delta kL}{\frac{1}{2}\Delta kL} \right)^2 \\ &= 2 \left(\frac{\mu_0}{\epsilon_0} \right)^{3/2} \frac{\omega^2 d^2}{\eta^2(\omega)\eta(2\omega)} I_\omega^2(0) L^2 \left(\frac{\sin\frac{1}{2}\Delta kL}{\frac{1}{2}\Delta kL} \right)^2. \end{aligned} \quad (3.28)$$

This Equation gives the intensity of the second-harmonic wave under the approximation that the fundamental wave is unattenuated in the medium, and as expected, the intensity of the second-harmonic wave is seen to increase as the square of the fundamental wave. Since the phase velocities of the fundamental and second-harmonic wave is different, the second-harmonic wave do not receive enough energy from the fundamental wave so the approximation that the wave is unattenuated is fair at this point.

From this we can now obtain the power conversion efficiency for second-harmonic generation, C_{SHG} as

$$C_{SHG} = \frac{I_{2\omega}(L)}{I_\omega(0)} \quad (3.29)$$

$$= 2 \left(\frac{\mu_0}{\epsilon_0} \right)^{3/2} \frac{\omega^2 d^2}{\eta^2(\omega)\eta(2\omega)} I_\omega(0) L^2 \left(\frac{\sin\frac{1}{2}\Delta kL}{\frac{1}{2}\Delta kL} \right)^2. \quad (3.30)$$

We see in Equation (3.30) that the conversion efficiency of the second-harmonic generation is maximised when $\Delta k = 0$ or when L is very small. This leads to the phase matching of making $\Delta k = 0$. When phase matching is achieved, the conversion efficiency of the second-harmonic generation increases, in other words, there is transfer of energy from the fundamental to the second-harmonic wave and we can no longer neglect the depletion of the fundamental wave. As a result of this depletion, the second-harmonic generation reaches a saturation point where it maintains a constant value, this occurs because the source of its energy which is from the fundamental wave is depleted.

In an experiment, using BBO (barium borate) as the nonlinear crystal for generating a second-harmonic signal, the relation between the average input power and the generated second-harmonic signal is illustrated in Figure 3.1. In this experiment there was phase matching. A quadratic polynomial was fitted into the data for the input power versus the second-harmonic signal, the fitting on the curve showed that for small signals the the intensity of the second-harmonic wave increases quadratically as the fundamental or pump wave. This experimental result confirms our theoretical calculations as expressed in equation (3.28).

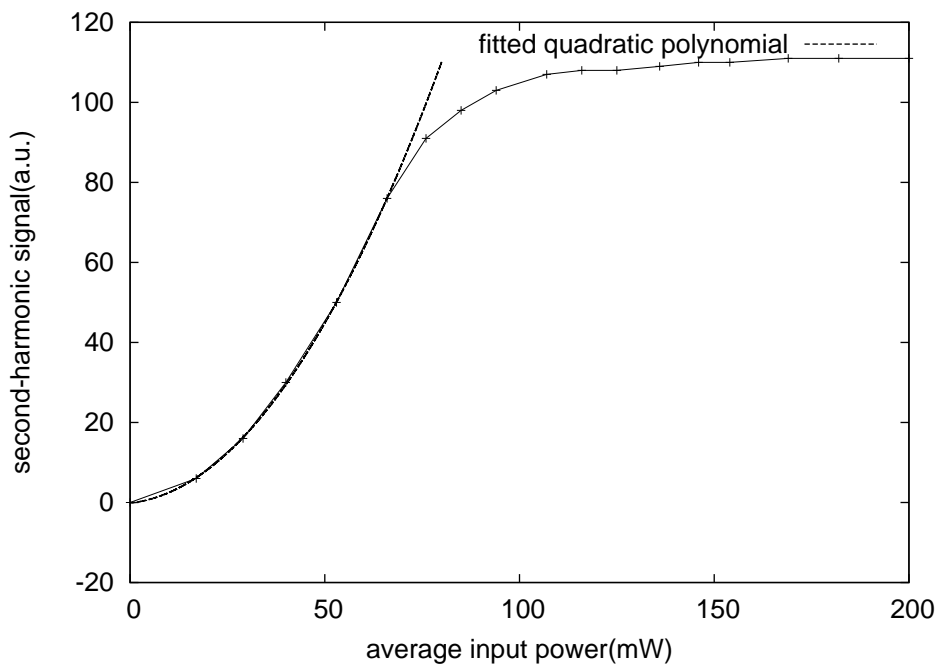


Figure 3.1: Intensity of the second-harmonic signal versus the average input power

However, in cases where the length of the crystal is small we do not bother about phase matching because length of the crystal is within the coherence length. We will discuss the coherence length in section 3.2. When $\eta(\omega)=\eta(2\omega)$, this means there is phase matching ($\Delta k = 0$) as in Equation (3.17), and

$$\lim_{x \rightarrow 0} \frac{\sin^2 x}{x^2} = 1 \implies \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L} \right)^2 = 1. \quad (3.31)$$

Thus in this case, the conversion efficiency

$$C_{SHG} = 2 \left(\frac{\mu_0}{\epsilon_0} \right)^{3/2} \frac{\omega^2 d^2}{\eta^3} I_\omega(0) L^2, \quad (3.32)$$

for $\eta = \eta(\omega) = \eta(2\omega)$.

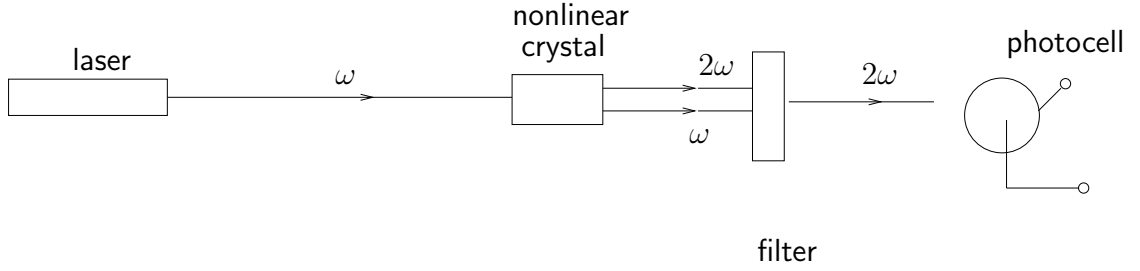


Figure 3.2: Schematic experimental arrangement for detection of second-harmonic generation, where ω is the frequency of the incident laser beam.

3.2 Phase Matching Condition

Phase matching is an important aspect of second-harmonic generation and nonlinear optics in general. The phenomenon of phase matching in relation to second-harmonic generation can be explained thus:

the factor ΔkL in Equation (3.28) determines the phase mismatch factor $e^{(2ik_\omega z - ik_{2\omega} z)}$ (as expressed in Equation (3.13)) between the fundamental and second-harmonic waves over the distance $z = L$. This is as a result of the different refractive indexes for the two waves.

The reason for the phase mismatch ΔkL between the fundamental (pump) and second-harmonic fields can be explained as described below: the second-harmonic field propagates with the phase velocity $\frac{c}{\eta(2\omega)}$, but we can see that from Equation (3.7) the nonlinear polarisation source has a phase velocity $\frac{c}{\eta(\omega)}$. Because of this difference the two fields get out of step of each other. Since the medium will only allow the second-harmonic field to propagate at a phase velocity of $\frac{c}{\eta(2\omega)}$, there is a reduction in the second-harmonic generation determined by $\eta(2\omega) - \eta(\omega)$, and this reduction is expressed quantitatively by the factor $\left(\frac{\sin \frac{1}{2} \Delta kL}{\frac{1}{2} \Delta kL} \right)^2$. Taking a range of values $-2\pi \leq \frac{1}{2} \Delta kL \leq 2\pi$ (see Appendix B.1 for code), the function

$$f(L) = \left(\frac{\sin \frac{1}{2} \Delta kL}{\frac{1}{2} \Delta kL} \right)^2$$

was plotted as shown in Figure 3.3.

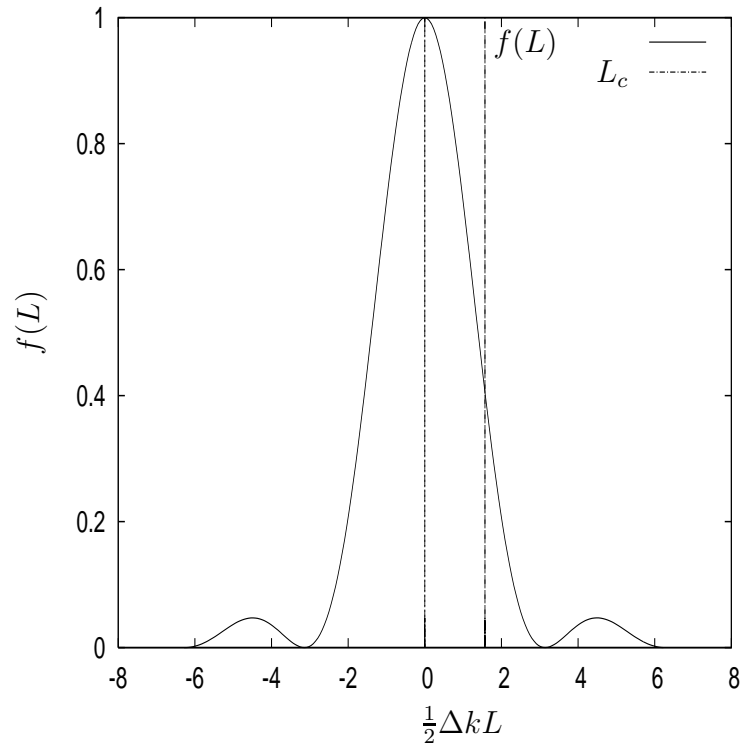


Figure 3.3: The graph of $f(L) = \frac{(\sin^2 \alpha L)}{(\alpha L)^2}$, where $\alpha \equiv \Delta k/2$.

The function drops to about 40 percent of its peak value at the distance

$$\frac{1}{2}\Delta k L = \frac{\pi}{2},$$

or

$$\Delta k L = \pi.$$

Therefore, we define the coherence length L_c , such that

$$\Delta k L_c = \pi,$$

or

$$L_c = \left| \frac{\pi}{\Delta k} \right|.$$

The coherence length is a certain fraction of L over which there is a significant second-harmonic generation. If there is a perfect phase matching then L_c is large, that is $\Delta k \rightarrow 0$. The conversion efficiency C_{SHG} as expressed in Equation (3.30), increases as the square of the length L . When phase matching is not achieved, however, only the coherence length L_c determines the conversion efficiency. If phase matching is not achieved, increasing L beyond L_c does not increase the conversion efficiency because the coherence length is usually very small on the order of $10\mu\text{m}$ [4].

Phase matching can be achieved in different ways, That is, making $|\Delta k|$ small as possible. One common method involves using *birefringent uniaxial crystals* to phase match the incident and second-harmonic field. We would briefly discuss what a birefringent material is so that we understand why it is useful in phase matching.

3.2.1 Birefringence

Let us discuss the birefringent nature of anisotropic materials as this will help us understand how and why they are used for phase matching. We have assumed that the elastic force acting on a bound electron is independent of the electron displacement \mathbf{r} . In general, however, “the spring constant” k may depend on the direction of \mathbf{r} , in which case the medium is said to be optically anisotropic. In such a medium the refractive index will be different for different polarisation of the field propagating through the medium. This is called birefringence [4].

We can understand how anisotropy may arise when we consider a long rod-shaped molecule in which the elastic restoring force is different for electron displacement parallel and perpendicular to any axis of the molecule. There will be different refractive indices for different direction of field polarisation in a medium in which there is some degree of molecular alignment. Most birefringent materials are uniaxial in the sense that they have one *optical axis*. An optical axis is one in which the refractive index is independent of the direction of the polarisation of the medium.

3.2.2 Angle Phase Matching Method

The method involves achieving a particular orientation of a birefringent crystal with respect to the propagation direction of the incident light. This method is most easily described if the crystal is uniaxial, that is, if it has only one optical axis, so we will consider the case where the crystal is uniaxial. Only two types of waves can propagate in a birefringent uniaxial crystal namely:

1. Waves that are polarised in a direction that is perpendicular to the plane of the optical axis $\hat{\mathbf{c}}$ of the crystal and the direction of propagation \mathbf{k} . Such waves are called *ordinary waves* and their refractive index η_o is called ordinary refractive index.
2. Waves polarised in a direction parallel to the plane of the optical axis and the direction of propagation. The waves are called *extraordinary waves* and their refractive index $\eta_e(\theta)$ which depends on the angle θ between the optic axis and the propagation vector \mathbf{k} , is called extraordinary refractive index,

The extraordinary wave with $\eta_e(\theta)$ depends upon the angle θ between the optic axis and \mathbf{k} by the relation[3]

$$\frac{1}{\eta_e^2(\theta, \omega)} = \frac{\sin^2 \theta}{\eta_e^2(\omega)} + \frac{\cos^2 \theta}{\eta_o^2(\omega)}. \quad (3.33)$$

Here, $\eta_e(\omega)$ is the value of $\eta_e(\theta, \omega)$ for $\theta = 90$ degrees and is equal to η_o for $\theta = 0$. The angular dependence of the extraordinary ray expressed in Equation (3.33) is as a result of the geometry of the nonlinear crystal, for this work we will not look at its derivation.

Phase matching is achieved by adjusting the angle θ to achieve a value of $\eta_e(\theta, \omega)$ for which the condition $\Delta k = 0$ is satisfied.

so we can deduce that

$$\eta_e(\omega) > \eta_0$$

is necessary to satisfy the requirement that $\sin^2 \theta_p > 0$. Note that if $\sin^2 \theta_p = 0$, then $\theta_p = 0$ and we do not want $\theta_p = 0$, else $\eta_e(\omega) = \eta_0$ and this will not help since we are considering a positive uniaxial crystal to help phase match the pump and the second-harmonic wave. Also we see that $\eta_0(\omega)^{-2} - \eta_0(2\omega)^{-2} < \eta_0(\omega)^{-2} - \eta_e(\omega)^{-2}$ which implies

$$\eta_e(\omega) > \eta_0(2\omega) \quad (3.37)$$

is necessary for $\sin^2 \theta_p < 1$, meaning that θ_p is less than 90 degrees for a positive uniaxial crystal. So we can conclude that, this method of phase matching is used in a positive uniaxial crystal when the condition in Equation (3.37) has been met, since $\eta_0(2\omega) > \eta_0(\omega)$ and Equation (3.37) both imply that $\eta_e(\omega) > \eta_0(\omega)$.

In practice the pump or fundamental wave is sent into the crystal as an extraordinary wave at the angle θ_p with respect to the optical axis and the second-harmonic wave is automatically generated as an ordinary wave propagating in the direction θ_p , because this is the only direction and polarisation for which there is phase matching and substantial second-harmonic generation [4].

For a negative uniaxial crystal the situation is reversed, phase matching is achieved if θ_p is chosen such that

$$\eta_e(2\omega, \theta_p) = \eta_0(\omega).$$

We chose the fundamental wave to be an ordinary wave and the second-harmonic wave to be an extraordinary wave. The angle between the pump and fundamental wave is derived in a similar manner as in Equation (3.33)

$$\frac{1}{\eta_e^2(\theta_p, 2\omega)} = \frac{\cos^2 \theta_p}{\eta_0^2(2\omega)} + \frac{\sin^2 \theta_p}{\eta_e^2(2\omega)}, \quad (3.38)$$

Solving for $\sin^2 \theta_p$

$$\sin^2 \theta_p = \frac{\eta_0(2\omega)^{-2} - \eta_0(\omega)^{-2}}{\eta_0(2\omega)^{-2} - \eta_e(2\omega)^{-2}},$$

and this can be written as

$$\sin^2 \theta_p = \frac{\eta_0(\omega)^{-2} - \eta_0(2\omega)^{-2}}{\eta_e(2\omega)^{-2} - \eta_0(2\omega)^{-2}}. \quad (3.39)$$

Since

$$\eta_0(\omega)^{-2} - \eta_0(2\omega)^{-2} < \eta_e(2\omega)^{-2} - \eta_0(2\omega)^{-2},$$

is required, then phase matching may be achieved if

$$\eta_e(2\omega) < \eta_o(\omega) \text{ (negative uniaxial)}. \quad (3.40)$$

4. Conclusion

Using the electron oscillation model, the linear and nonlinear response of an electron to an applied optical field was modelled. The nonlinear response of the electrons to the applied force showed that the polarisation of a material can be expressed as a power series in the applied electric field, such that for light with high intensities such as laser light, frequencies, doubling that of the incident light can be obtained. As a result of this, we solved Maxwell's equation and observed a nonlinear source term $\mathbf{P}^{NL}(\omega)$ that could generate coherent radiation at a new frequency. So treating the case of second-harmonic generation, we considered the nonlinear polarisation source of $P(2\omega)$ that could generate an electric field propagating at the frequency 2ω . The following deductions were made on the efficiency of the generation of a second-harmonic generation:

1. The intensity of the second-harmonic signal depends on the square of the incident laser light under the approximation that the fundamental wave is unattenuated inside the medium. In practice however, the fundamental wave becomes depleted as it passes through the medium because its energy is been used to generate the second-harmonic wave. Experimental results showed that the effect of this depletion is a saturation in the generation of the second-harmonic signal, from which that the second-harmonic signal maintains a constant value.
2. The conversion efficiency of the second-harmonic generation depends on the square of the second order nonlinear susceptibility $\chi^{(2)}(2\omega, \omega, \omega)$ which is enhanced near the resonance frequency. So with tunable lasers we can tune the frequency of the laser into close resonance with atomic transition frequencies[1].
3. The generation of the second-harmonic wave was limited by a phase mismatch factor which arose from the fact that the speeds of the fundamental and second-harmonic wave are different and this difference could reduce the conversion efficiency of the second-harmonic wave. In order to compensate for this reduction, it was shown that the crystal for second-harmonic generation should be birefringent so that they could be phase matching between the fundamental and second-harmonic wave by the orientation of the crystal at an angle θ_p . This angle of orientation was calculated for positive and negative uniaxial crystals.

In any future work on this topic, I recommend that the angular dependence of the nonlinear susceptibility tensor $\chi^{(2)}$ be studied.

Appendix A. Code for the computation of the variation of the refractive index with frequency

A.1 A code for the dispersion curve

This code computes the variation of the real part of refractive index with increasing frequency.(see section 1.5)

```
from __future__ import division
import Gnuplot
from scipy import *
g = Gnuplot.Gnuplot(persist=1)

C = 1.
w0 = 20.
gamma = 2.
index = []
lst = []
for w in arange(0,40,0.005):

    n = 1+ C*(w0**2-w**2)/((w0**2-w**2)**2 + w0**2*gamma**2)
    index.append([w,n])
    lst.append([w0,n])

data2 = Gnuplot.Data(lst, with = 'lines 30')
data = Gnuplot.Data(index, with = 'lines ')
g('set xlabel "frequency"')
g('set ylabel "refractive index"')
g.plot(data,data2,title = 'Real part of refractive index')
g.hardcopy(filename="realpart.eps",terminal="postscript",mode = "landscape")
```

A.2 A code for the imaginary part of the refractive index

This code computes the variation of the imaginary part of refractive index with increasing frequency.(see section 1.5)

```
from __future__ import division
import Gnuplot
from scipy import *
g = Gnuplot.Gnuplot(persist=1)

C = 1
w0 = 20.
gamma = 2.
index = []
lst = []
for w in arange(0,40,0.005):

    I = C*((w*gamma)/((w0**2 -w**2)**2 + w**2*gamma**2))
    index.append([w,I])
    lst.append([w0,I])

data2 = Gnuplot.Data(lst, with = 'lines 30')
data = Gnuplot.Data(index, with = 'lines ')
g('set xlabel "frequency"')
g('set ylabel "refractive index"')
g.plot(data,data2,title = 'Imaginary part of refractive index')
g.hardcopy(filename="imaginarypart.eps",terminal="postscript",mode = "landscape")
```

Appendix B. Code to Compute Reduction in the Second Harmonic Generation and Coherence Length

B.1 Coherence length

```
from __future__ import division
from scipy import *
import Gnuplot

g = Gnuplot.Gnuplot(persist = 1)

g('set xlabel "L"')
g('set ylabel "f(L)"')
data = []
data1=[]
data2 = []
data3 = []
y = pi/2
z=0
for t in arange(-2*pi, 2*pi, pi/12):
    f = sin(t)**2/(t**2)
    if t <= pi/2:
        print t, f
        data.append([t, f])
        data1.append(f)
        data2.append([y,f])
        data3.append([z,f])

print max(data1)
plot1 = Gnuplot.PlotItems.Data(data, with = 'lines', title = 'victoria is a good girl')
plot2= Gnuplot.PlotItems.Data(data2,with = 'lines 23',title='LC' )
plot3= Gnuplot.PlotItems.Data(data3,with = 'lines 23', )
g.plot(plot1,plot2,plot3)

g.hardcopy(filename="phasemact.eps",terminal="postscript")}
```

Appendix C. Code for plotting the Intensity of the Second-Harmonic Signal

C.1 Fitted Quadratic Polynomial on the Generation of the Second-Harmonic Signal

```
from __future__ import division
from scipy import *
from pylab import *
import Gnuplot
from scipy.io import *
from scipy.interpolate import *

g = Gnuplot.Gnuplot(persist = 1)
g('set xlabel "average input power(mW)"')
g('set ylabel "second-harmonic signal(a.u.)"')
#power=asarray(array_import.read_array("/home/victoria/Desktop/power.txt"),Int)
power = array([[0,0], [17,6], [29,16], [40,30], [53,50], [66,76], [76,91], [85,98], [94,103],
#power2=asarray(array_import.read_array("/home/victoria/Desktop/power2.txt"),Int)

plot1=Gnuplot.PlotItems.Data(power, with='lines')
plot4=Gnuplot.PlotItems.Data(power, with='points')
#plot3=Gnuplot.PlotItems.Data(power2, with='lines')

lst = []
data2=[]
for i in xrange(2): lst.append(list(power[:,i]))

t = range(len(lst[0]))
Xtck = splrep(t,lst[0],s=0)
Ytck = splrep(t,lst[1],s=0)
xspline=splev(arange(0.,20.1,0.01),Xtck,der=0)
yspline= splev(arange(0.,20.1,0.01),Ytck,der=0)

k = arange(0.0,80.,0.001)
Yfit = polyfit(lst[0][0:6],lst[1][0:6],2)
#Yfit = polyfit(t,lst[1],2)
#xx = polyval(Xfit, arange(0.0,10.,0.001))
yy = polyval(Yfit, arange(0.0,80.,0.001))
#plot(k,yy)
```

```
for h in xrange(len(k)):
    data2.append([k[h],yy[h]])

plot2 = Gnuplot.PlotItems.Data(data2,with = 'lines lw 2',title='fitted quadratic poly

g.plot(plot1,plot2,plot4)

g.hardcopy(filename="power.eps",terminal="postscript",fontsize=20)
```


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