A RUDIMENTARY VIEW ON SECOND HARMONIC GENERATION

V.S.Kumar [*]

R.S.Sundararajan [**]

Abstract :

The enormity of nonlinear optic materials apostasy in the validity that the second harmonic generation is one of the prime methods of efficacious conversion of coherence light rays. Many such characterized materials have been utilized in optoelectronic contrivances for sundry purposes like data storage, data conversion and switching mechanisms. This article expresses the scrutiny on the second harmonic generation and its purport theoretically such as frequency matching, phase matching, amplification, wave commixing and their modalities.

Keywords : Nonlinear optic, Second harmonic generation, Phase matching, Frequency conversion, Non centrosymmetric.

[*] **Department of Physics, Swami Dayananda College of Arts and Science, Manjakkudi, Tiruvarur Dt.,Tamilnadu, India.**

[**] **Department of Physics, Government Arts College(Autonomous), Kumbakonam, Tamilnadu, India.**

1. Introduction:

The development of nonlinear optical materials has been driven by a multitude of consequential technological applications that can be realized if congruous materials are available. Future generations of optoelectronic contrivances for telecommunication, information storage, optical switching, and signal processing are soothsaid to an astronomically immense degree on the development of materials with exceptional nonlinear replications [1-4].

Under mundane conditions the replication of a medium to light is linear and as a consequence most optical phenomena can be described with a linear refractive index. It was only with the invention of lasers in the early 1960's that the available optical power level incremented to a caliber where the replication of the medium commenced to deviate from the linear demeanor. It was found that at sufficiently high light intensities the replication of the material depends on the light intensity. This led to an entirely incipient field of nonlinear optics and the revelation of sundry intriguing phenomena such as second harmonic and sum - difference frequency generation [5]. Nowadays, frequency conversion is a common tool to create blue or infrared laser light in the laboratory; i.e. at frequencies where no alternative laser sources are available.

1.1. High degree coherence source:

Usually, the field vitality of the conventional light sources used afore laser invention were more minuscule than the field strengths of atomic fields. Because there transmutation is of the order of mV per cm and above. High degree of coherence of the laser radiation have prodigiously spatial concentration of light power to engender 1 MW pulses even lasting a few tenths of ns. At such fields, the cognation between polarization and energy ceases to be linear. Bloembergen established theoretical wave for NLO. NLO properties have been demonstrated by harmonic generation of light firstly observed by Franken and his coworkers in 1961 [6]. They observed UV light at twice the frequency of ruby laser, when the light was made to negotiate a quartz crystal. This attracted wide spread for NLO investigations.

2. Experiment : A ruby laser beam of order 10 kW focused on a quartz slab. The transmitted light passed through a filter which cuts off the red light and allows UV light to pass through; it incident on a photocell with power 1MW was then observed.(Figure.1)

Figure.1 Ruby laser experiment

3. Second harmonic generation :

A sizably voluminous number of organic Π conjugated molecules have been investigated in the last three decades for sustainability function as components in hypothetical NLO materials. All NLO effect waves were not discovered until the revelation of laser. Parametric amplification was observed in $LiNbO_3$, by two wave commixing in temperature tuning single crystals[7].

Second harmonic generation was first realized by quartz and generated in many other crystals like KDP, ADP and Barium nitrate. SHG is also realized in gases and semiconductors[8,9].

3.1. Theory :

Dielectric medium when placed in electric field, polarization occurs, if the medium does not have a transition at the frequency of the field. Each molecule acts like dipole with its moment p_i.

$P = \Sigma_i p_i$

Σ_i is the summation over dipoles per unit volume. Compass reading effect of the field E on dipole depends on the propagating medium and strength.

$P = \varepsilon_0 \chi E$

where χ is the polarizing susceptibility and it does not depends on the field E.

Dielectric permeability(μ) depends on χ and E.

$\mu \alpha \chi$

and

$\mu \alpha E$.

Consider the field incident on a medium,

$$E = E_0 \text{Cos}\omega t \quad \text{----------(1)}$$

Therefore, $P = \varepsilon_0 E_0 \chi^1 \text{Cos}\ \omega t + \varepsilon_0 E_0 \chi^2 \text{Cos}^2\omega t + \varepsilon_0 E_0 \chi^3 \text{Cos}^3\omega t + \quad \text{----------(2)}$

The first term give rise to dc field. The second term give rise to first harmonic polarization, the next term refers second harmonic and the last term represents the third harmonic polarization.

$$P = \tfrac{1}{2}\varepsilon_0 E_0^2 \chi^2 + \varepsilon_0(\chi^1 + \tfrac{3}{4}\chi^3 E_0^2) E_0 \cos\omega t + \tfrac{1}{2}\varepsilon_0 E_0^2 \chi^2 \cos 2\omega t + \tfrac{1}{4}\varepsilon_0 E_0^3 \chi^3 \cos 3\omega t + \text{------} \quad \text{----------(3)}$$

A polarization oscillating at frequency 2ω radiates as electromagnetic wave of the same frequency which propagates with the same velocity as that of the incident wave with same characteristics of directionality and monochromacity as the incident wave. Nonlinear polarization depends on the propagation direction, electric field polarization and orientation of optic axis.

$$P_i^{\,2} = \varepsilon_0 \Sigma_{jk} \chi_{ijk}^{\,2} E_j E_k \quad \text{----------(4)}$$

i,j,k are the coordinates of x,y,z axes. If crystals are isotropic χ_{ijk} is independent on direction. Second harmonic generation cannot occur in isotropic medium nor in centrosymmetric crystals; only crystals lack inversion symmetric exhibits this. In case of uniaxial crystals both the quadratic and cubic forms are present.

Let we hold that,

$$P_{nl} = 2d\varepsilon^2 \quad \text{----------(5)}$$

here ε the electric field comprising one or two harmonic components and determine the spectral components of P_{nl}. With the first born approximation, the radiation source contains the same spectral components as P_{nl} and therefore does the sprinkled field.

3.2. Optical adaptation:

Consider the response of this non linear medium to a harmonic electric field of angular frequency ω and complex amplitude E(ω),

$$\varepsilon(t) = \mathrm{Re}\{ E(\omega)\exp(j\omega t)\} = \tfrac{1}{2}[E(\omega)\exp(j\omega t) + E^*(\omega)\exp(-j\omega t)]$$

----------(6)

The non linear polarization density P_{nl} is given by $P_{nl}(t)$

(on substituting.eqn.5)

$$P_{nl}(0) + \mathrm{Re}\{P_{nl}(2\omega)\exp(j\omega t)\} \quad \text{----------(7)}$$

$$\text{Since, } P_{nl}(0) = d\,E(\omega)E^*(\omega) \quad \text{----------(8)}$$

$$P_{nl}(2\omega) = d\,E^2(\omega) \quad \text{----------(9)}$$

The component $P_{nl}(0)$in eqn.(7) corresponds to a firm polarization density which creates a potential difference in dc across the plates of a capacitor within which the nonlinear material is located. The generation of a dc voltage as a result of an intense optical field represents optical rectification.

The source $S(t) = -\mu_o \partial^2 P_{nl}/\partial t^2$ corresponding to eqn. (7) has a component at 2ω with amplitude $S(2\omega) = 4\mu_o \omega^2\, dE(\omega)E(\omega)$ which radiates an optical field at 2ω . Thus the scattered optical field has a component at the second harmonic of the incident optical field. The amplitude is proportional to S(2ω), its intensity I(2ω) is proportional to $|S(2\omega)|^2$ which is proportional to the square of the intensity of the incident wave $I(\omega) = |E(\omega)|^2 / 2\eta$ and to the square of the non linear coefficient d.

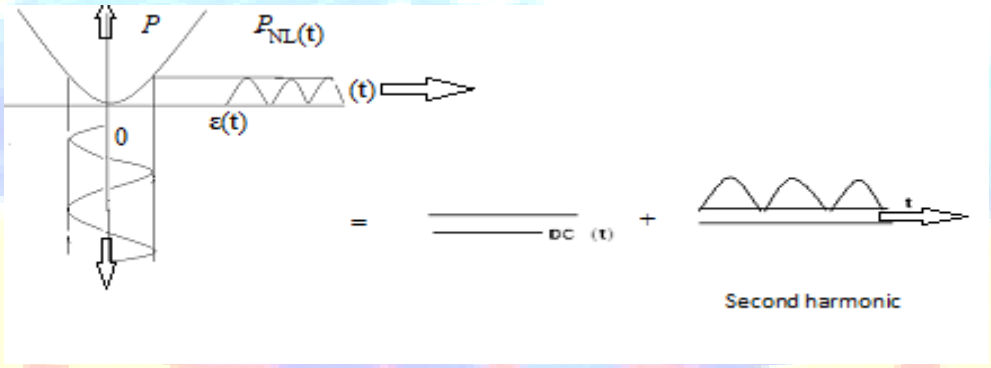

Figure.2 A sinusoidal electric field in a second-order nonlinear optical medium

The efficiency is given by $\eta = C^2 L^2 P / A$. C^2 depends on d^2 and ω^2. To exploit the efficiency, it is essential that the incident wave have the most sizably voluminous possible power P. This is proficient by consumption of pulsed lasers. If the dimensions of the non linear crystal are not circumscribing factors the maximum value of L for a given area A is circumscribed by beam diffraction. For a thick crystal, the beam should be focused to the most sizably capacious spot that fits within the cross-sectional area of the crystal. For a thin crystal, L is tenacious by the crystal and the beam should be focused to the most infinitesimal spot area.

3.3. Three wave mixing :

The second order non linear medium can be used to mix two optical waves of different frequencies and generate a third wave at the different frequency or at the sum frequency. Although the incident pair of waves at frequencies produce polarization densities at different frequencies, these waves are not necessarily generated, since phase matching must be satisfied. Phase matching condition is also important in frequency mixing. In this case, it is necessary to find a direction in a crystal.

3.4. Modalities :

When two optical waves of angular frequencies ω_1 and ω_2 peregrinate through a second order nonlinear optical medium they commix and bring about a polarization density with components at a number of frequencies. We postulate that only the component at the sum frequency $\omega_3 = \omega_1+\omega_2$ slakes the phase-matching condition. Other frequencies cannot be sustained by the medium since they are surmised not to slake the phase matching condition. Once wave 3 is engendered, it interacts with wave 1 and engenders a wave at the difference frequency $\omega_2 = \omega_3 - \omega_1$. Limpidly, the phase-matching condition for this interaction is additionally gratified. Waves 3 and 2 similarly coalesce and radiate at ω_1. The three waves consequently undergo mutual coupling in which each pair of waves interacts and contributes to the third wave. The process is called three-wave commixing. Two-wave commixing is not, in general, possible. Two waves of arbitrary frequencies ω_1 and ω_2 cannot be coupled by the medium without the avail of a third wave. Two wave commixing can occur only in the degenerate case, $\omega_2 = 2\omega_1$, in which the second harmonic of wave 1 contributes to wave 2; and the sub harmonic $\omega_2 / 2$ of wave 2, which is at the frequency difference $\omega_2 - \omega_1$, contributes to wave 1. Three-wave commixing is kenned as a parametric interaction process.

i. Optical Frequency Conversion (OFC): Waves 1 and 2 are mixed in an **up converter**, generating a wave at the sum frequency $\omega_3 = \omega_1 + \omega_2$. This process, also called **sum-frequency generation**.(Figure.3a)

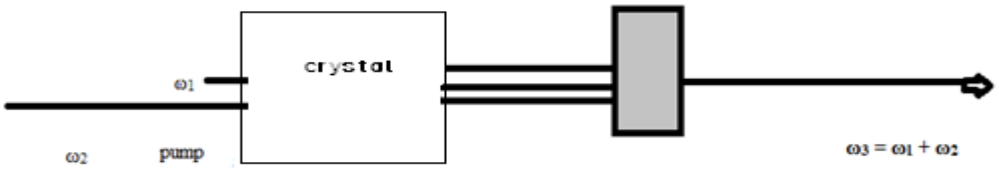

Figure.3a Optical frequency converter

ii. Second-harmonic generation is a deteriorate of SFG. The opposite process of **down conversion** is realized by an interaction between waves 3 and 1 to generate wave 2, at the difference frequency $\omega_2 = \omega_3 - \omega_1$. Up and down-converters are habituated to create coherent light at wavelengths where no ample lasers are available.

iii. Optical Parametric Amplifier (OPA): Waves 1 and 3 interact, so that wave 1 grows, and in the process wave 2 is created. The device operates as a coherent amplifier at frequency ω_1 and is known as an OPA. Wave 3, provides the required energy. The amplified wave is called the **signal**. Undoubtedly, the gain of the amplifier depends on the power of the pump. These are used as sensitive detectors.(Figure.3b)

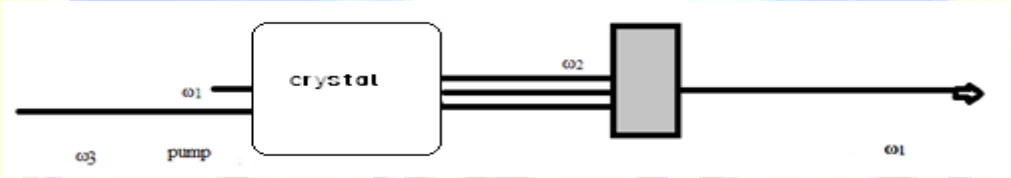

Figure.3b Optical parametric amplifier

iv. Optical Parametric Oscillator (OPO): The parametric amplifier as an oscillator, in which pump wave is supplied and are used for the generation of coherent light and mode-locked pulse trains in bands.

v. Spontaneous Parametric Down conversion (SPDC): The input to the nonlinear crystal is wave 3, and down conversion to the lower-frequency waves 2 and 3 is impulsive. The phase-matching conditions lead to formation of pair of waves 1 and 2 with specific frequencies and directions.

3.5. Phase matching:

Phase matching criterion becomes a refractive index criterion. Most materials show some scarcely dispersion in the refractive index. A birefringent material has different refractive indices for different polarization due to low symmetry crystals. The uniaxial crystal waves whose refractive index is independent of the propagation direction, called as ordinary ray; which depends on it refers to extraordinary ray. Indicatrix for a crystal for ordinary type refers to spherical and extraordinary refers to ellipsoid.

The intensity of the exiting radiation controls the harmonic generation efficiency and the direction of propagation in crystals. Suppose the second harmonic wave at 2ω driven by it are propagating in the z direction through a material. The amount of second harmonic radiation produced within a portion of width located will be proportional to the width and the second harmonic dipole moment per unit volume induced at frequency 2ω.

3.6. Choosing a material:

Refractive index for extra ordinary ray at 2ω is equal to the ordinary ray at ω. Then only effective free conversion is possible.

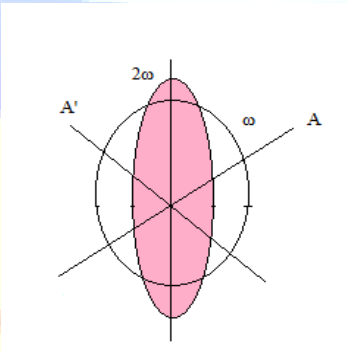

Figure.4 Refractive index surfaces of a uniaxial crystal

Ordinary wave refractive index and extra ordinary intersect at A(Figure.4)

$\eta_o\omega = \eta_e 2\omega$

Thus second harmonic propagation in this direction is phase matched.

4. Conclusion :

The magnitude of second harmonic generation deceit in the authenticity that it is one of the principal methods of efficacious conversion of infrared radiation into visible and visible into ultraviolet. Many non linear materials have been characterized by SHG method and the culled felicitous materials are utilized in optoelectronic contrivances for sundry purposes like storage, conversion and switching mechanisms. Astonishing advances have been made by applying this method for all non-centrosymmetric crystals.

References :

1. Zyss J, editor. Molecular Nonlinear Optics: Materials, Physics and Devices. New York: Academic Press; 1994.

2. Chemla DS, Zyss J, editors. Nonlinear Optical Properties of Organic Molecules and Crystals. Vols. 1 and 2. Orlando, FL: Academic Press; 1987.

3. Prasad PN, Williams D, editors. Introduction to Nonlinear Effects in Molecules and Polymers. New York: J. Wiley; 1991.

4. Papadopoulos MG, Leszczynski J, Sadlej AJ, editors. Nonlinear Optical Properties of Matter: From Molecules to Condensed Phases. Dordrecht: Kluwer; 2006.

5. Y.R. Shen, *The principles of nonlinear optics* (John Wiley, New York, 1984).

6. Franken, P.A., Hill, A.E., Peters, C.W. and Weinrich, G.Phys.Rev.Lett.7,118(1961).

7. Goirdmaine A, Miller RC. Phys. Rev. Lett. 1965;14:973.

8. A D White and J D Rigden, Proc. IRE 50, 1697 (1962).

9. Armstrong, J.A., Nathan, M.I. and Smith, A.W., App. Phys. Lett.3,68(1963)