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THEORIES BEHIND NON LINEAR OPTICS

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ABSTRACT

Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light. Nonlinear optical phenomena are "nonlinear" in the sense that they occur when the response of a material system to an applied field depends in a nonlinear manner on the strength of the optical field. Typically, only laser light is sufficiently intense to modify the optical properties of a material system. In nonlinear optics, superposition principle no longer holds. The beginning of the field of nonlinear optics is often taken to be the discovery of second harmonic generation by Franken et al. shortly after the demonstration of the first working laser by Maiman in 1960.

Nonlinear optics provides the basis of many key technologies used today for providing radiation sources in various wavelength ranges from X-rays to the tetrahertz. Nonlinear optical technologies are usually used in combination with laser technology, and the main aspect is to shift or extend the limited wavelength range directly accessible by the laser source.

KEYWORDS: *Interatomic, Laser, Linear Optics, Nonlinear Optics, Intensity, Field Strength*

INTRODUCTION

Light is a prima donna in the natures opera! Signifying knowledge and life, light played the crucial role, when life evolved on the Mother Earth. Linear optics is a sub-field of optics, consisting of linear systems and it includes most applications of lenses, mirrors, wave plates, diffraction gratings and many other common optical components. Linear optics has the following properties- i)if monochromatic light enters an unchanging linear optical system, the output will be at the same frequency ii) superposition principle is valid iii) repeatedly, if the input light is made more intense, then the output light is made more intense but otherwise unchanged.

When we use a relatively low light intensity that normally occur in nature and obtained from any light source, the optical properties are quite independent of the intensity of illumination. It was thought that all optical properties are linear and superposition principle, a fundamental tenet of classical physics is applicable.

On the other hand nonlinear optics is the branch of optics that describes the behaviour of light in nonlinear media, that is, media in which the polarisation density P responds non-linearly to the electric field strength of the light. The non linearity is typically observed only at very high light intensities (values of atomic electric fields, typically 10^8 V/cm) such as those provided by lasers. The behaviour is not observed when light travels in free space but resides in the medium through which light passes. The interaction of light with light is therefore mediated by the nonlinear medium,

Such behavior provides insight into the structure and properties of matter. Experiments carried out in the post-laser era clearly demonstrate that optical media do in fact exhibit nonlinear behavior, as exemplified by the following observations:

- The refractive index, and consequently the speed of light in a nonlinear optical medium, is dependent on light intensity.
- The principle of superposition is not valid
- The frequency of light is changed when it passes through a nonlinear optical medium
- Photons can be involved in interaction within the confines of a nonlinear optical medium so that light can indeed be used to control light.

The field of nonlinear optics offers a variety of fascinating phenomena, many of which are also eminently useful. The presence of an optical field modifies the properties of the medium, which in turn causes another optical field, or even the original field itself, to be modified. Non-linear optical (NLO) effects are analyzed by considering the response of the dielectric material at the atomic level to the electric field of intense light beam (laser).

HISTORICAL BACKGROUND OF NONLINEAR OPTICS

The first demonstration of Nonlinear Optical (NLO) frequency conversion took place as early as in 1961.It was done by Franken et al.[2]. They used quartz crystal to double the frequency of 694 nm light of Ruby laser. The theoretical underpinning for both microscopic origin of the nonlinear susceptibilities and the propagation effects governing macroscopic nonlinear interactions between electro-magnetic waves was discussed in the classical paper of Armstrong et al.[1]. Later on, Nicolas Bloembergen won Nobel Prize in 1981 for his contribution in laser spectroscopy. Also, the selected papers of Bloembergen were published under the title, 'Encounter in the Nonlinear Optics'[3]. Then the quest began for identifying various nonlinear optical materials.

First Demonstration of Second Harmonic Generation.

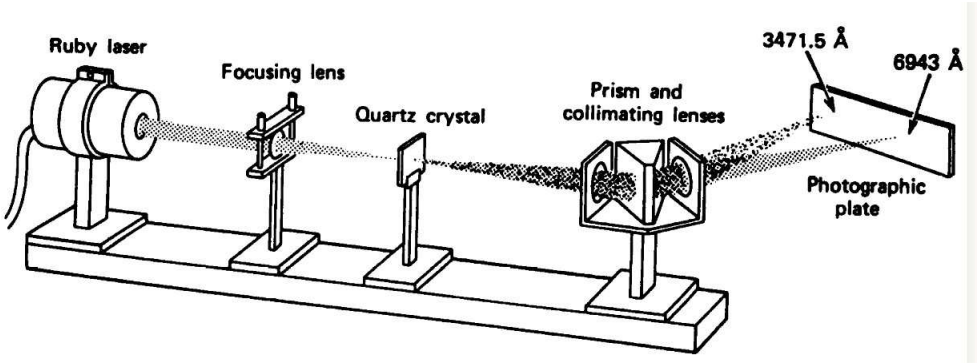

Figure 1: Arrangement used in the first experimental demonstration of Second harmonic generation [P. A. Franken, et al.[2]]. A ruby laser at λ = 0.694 μm is focused on a quartz crystal, causing the generation of a (weak) beam at λ/2 = 0.347 μm. The two beams are then separated by a prism and detected on a photographic plate.

DISCUSSION (ORIGIN OF NON-LINEAR OPTICS)

Nonlinear optics (NLO) is the study of interaction of intense electromagnetic field with materials .This interaction produces modified fields that are different from the input field in phase, frequency or amplitude[1,4-6]. The physics behind nonlinear optics is best understood when we concentrate on the effect of a field on the electrons of a nonlinear crystal. It is thought that electrons in a nonlinear crystal are bound in potential well, which acts like a spring, holding the electrons to lattice point in the crystal (**Fig. 2)**. When an external force is applied the force pulls an electron away from its equilibrium position. The spring pulls it back to its original position. The force is proportional to the displacement. The restoring force increases linearly with the electron displacement from its equilibrium position. The electric field in a light wave passing through the crystal exerts a force on the electrons and pulls them away from their equilibrium position. In an ordinary optical material, the electrons oscillate about their equilibrium position at the frequency of this electronic field. According to the fundamental law of physics an oscillating electron will radiate at its frequency of oscillation. These electrons in the crystal 'generate' light of the frequency of the original light wave. In linear materials, the response is always proportional to the stimulus. The induced polarization is proportional to the field and the susceptibility is independent of the field. In practice, this is always the case at low fields. On the contrary, in nonlinear materials electrons are thought to bound by very short springs. If highly intense light is passed through the material, its electric field can pull the electrons to far and they reach the end of their springs. The restoring force, here, is no longer proportional to the displacement .The substanceis then said to be non-linear. The electrons are jerked back roughly rather than pulled back smoothly. In that case, they oscillate at frequencies other than the driving frequency of the light wave. These electrons radiate at the new frequencies and generate new wavelength of light. The exact values of the new wavelengths are determined by conservation of energy. The energy of the new photon generated by the nonlinear interaction must be equal to the energy of the photons used. It is called Non-linear Optics because, at high intensity, the graph representing the dependence of optical polarization on the light field amplitude has curvature and deviates from straight line.

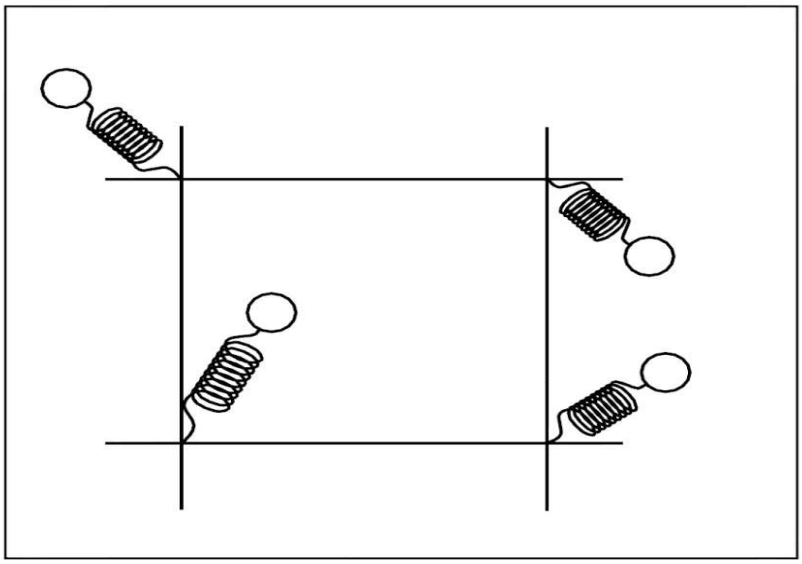

Figure 2: Electrons in a Nonlinear Crystal are Bound in a Potential Well, Holding the Electrons to Lattice Points.

Theoretical Background

Non linear optical materials are basically dielectric .When a dielectric medium placed in an electric field it is polarized, if the medium does not have a transition at the frequency of the field. In that case, each constituent molecule acts as a dipole, with a dipole moment, pi. Hence the dipole moment vector per unit volume, P, is given by P = ∑Pi where the summation is over the dipoles in the unit volume. The orienting effect of the external field on the molecular dipoles depends both on the properties of the medium and the field strength. Thus we can write,

$$P = €0\ \chi,\ E$$

Where χ is called the dielectric susceptibility of the medium. This relation is valid for the field strengths of conventional sources. The quantity χ, is a constant only in the sense of being independent of E; its magnitude is a function of the frequency. With sufficiently intense laser radiation, the relation does not hold good and has to be generalized to

$$P = €0(\chi^{1} E + \chi 2\ E^{2} + \chi 3\ E^{3} + ...)$$

Where χ1 is the same as χ, the coefficients χ2, χ3 define the degree of non-linearity and are known as non-linear susceptibilities. If the field is low, as it is in the case of ordinary light sources, only the first term can be obtained. It is for this reason that the pre-laser optics is known as linear optics. Higher the value of the electric field, more significant becomes the higher order terms. If we suppose that the field incident on a medium has the form

$$E = E0\ \cos(\omega t)$$

Then equation takes the form

$$P = €\ 0\ \chi 1 E_0\ \cos(\omega t) + €0\chi 2 E_0^{2}\ \cos 2(\omega t) + €_0\chi 3 E_0\ 3\cos 3(\omega t)$$

Here, the first term is a constant term. It gives rise to a dc field across the medium, refractive index, absorption, dispersion and birefringence of the medium. The second term follows the external polarization and is called the first or fundamental harmonic of polarization; the third oscillates at frequency, 2ω, and is called the second harmonic of polarization, the fourth is called the third harmonic of polarization. The optical field incident on the nonlinear optical medium consists of two distinct frequency components which gives rise to a number of nonlinear phenomenon in material medium, such as,

- Sum and difference frequency generation or Parametric generation,
- Optical rectification,
- Second harmonic generation.

When ω1 = ω2 = ω and, ω3 = 2ω, the polarization, P (ω3 = 2ω), is the source of second harmonic generation (SHG). When ω3 = 0, the effect is called optical rectification. These two results are special cases of the general process known as sum and difference frequency mixing where ω3 = ω1 + ω2. It can be seen that in ω3 = ω1 + ω2, if ω3 and ω1 are fixed, then ω2 is also fixed. If only ω3 is fixed, then the ω1 and ω2 are free to range over many values, this effect is known as parametric amplification. When ω1 = 0 and ω3 = ω2 = ω, then there is a change in the refractive index of a material by the presence of a dc or a low frequency ac field. The change depends linearly on the applied electric field strength. This is the Linear electro-optic (LEO) effect.

A noticeable SHG is observed only if all the SHG wavelets combine constructively along the propagation path of the incident wave. This is possible only if the fundamental (ω) and SHG (2 ω) waves are in phase (propagate in similar phase velocities), which is generally called being phase matched. However, due to normal dispersion of the most media where both ω and 2 ω waves travel at different phase velocities, these waves are in phase only for a distance of l_c where the intensity of the 2 ω wave become maximum. This distance is called coherence length, which is usually of the order of micrometer. Due to the short interaction length, such a non-phase matched conditions produces SHG several orders of magnitude lower than that generated under phase-matched conditions with an interaction length of several mm to cm. Phase matching condition can be achieved by using optically anisotropic (uniaxial or biaxial) NLO crystal.

Since χ2 is a second order polar tensor, it vanishes in a centrosymmetric environment. Therefore, most second order nonlinear optical materials have been composed so far from non-centrosymmetric one-dimensional charge transfer molecules. Typically they contain a conjugated pi electron system, asymmetrically substituted by electron donor and acceptor groups.

Optical rectification and the linear electrooptic effect (Pockels' effect) are nonlinear optical techniques for the generation and detection of freely propagating sub-picosecond THz-frequency radiation pulses. Generally, optical rectification refers to the development of a dc or low frequency polarization when intense laser beams propagate through a crystal. The linear electrooptic effect describes a change of polarization of a crystal from an applied electric field. Optical rectification and the linear electrooptic effect occur only in crystals that are not centrosymmetric. However, optical rectification of laser light by centrosym- metric crystals is possible if the symmetry is broken by a strong electric field. Furthermore, generation and detection of THz-radiation pulses by optical rectification and the Pockels effect require that the crystals are sufficiently transparent at THz and optical frequencies.

Freely propagating sub-picosecond THz-radiation pulses are generated by optical rectification of femtosecond (fs) near infrared laser pulses in crystals with appropriate nonlinear optical properties for this process. The detection of freely propagating THz-radiation pulses is performed by measuring the phase modulation of an fs near infrared laser pulse propagating through an electrooptic crystal simultaneously with a THz-radiation pulse. The electric field of the THz radiation induces a phase modulation of the fs laser pulse through the linear electrooptic effect.

Microscopic Nonlinearity

The optical nonlinearity of a medium is determined by the optical nonlinearities of the molecule. Therefore, it is important to understand the electronic origins of microscopic nonlinearity. The polarization of a molecule is often approximated as the creation of an induced dipole by an electric field. This dipole moment is given by

$$\mu i(E) = \mu 0 + \sum_j \alpha ij\ Ej + \sum_{jk} \beta ijk\ Ej\ Ek + \sum jkl \gamma ijkl Ej\ Ek\ El$$

Where μ0 is the permanent dipole moment. α is the polarizability of the molecule which describes the linear interaction with the optical field. β and γ are called the first and second hyperpolarizabilities and constitute the molecular origin of the second and third order nonlinear optical interactions. μ and β are non-zero only for noncentrosymmetric molecules. α and γ are non-zero for all molecules[7-10].

Resonant Molecular Optics

The large number of molecular structures gives rise to a large number of optical transitions (resonances). These resonances determines the response function to the electromagnetic fields and gives information about the nonlinear optical properties. There is enormous enhancement of the nonlinear signals when the incident electromagnetic field frequencies match the optical resonances of the structures. This leads to significant enhancement of the third order nonlinear susceptibility. Thus, the nonlinear optical properties of a material at resonant or near-resonant wavelengths are much greater than those operating at the off-resonant domain[11].

CONCLUSIONS

Non-linear optical effects of molecular crystals depends on the polarisability of the electrons in the pi bonding orbitals. The optical non-linearity can be increased by adding conjugated bonds (increasing the length of the conjugated system) or substituting donors(can donate electrons into the pi electron system) and acceptors (can accept electrons into the pi electron system). The addition of functionality at the ends of the pi system can enhance the asymmetric electronic distribution. Non-linear optical organic material can be incorporated in the form of polymer, thin film and crystals. The development of polymeric materials for SHG application has been less successful, in particular because of low non-linearities, absorption losses and phase-matching problems. The second approach is of LB films. LB films are often of poor optical quality due to microdomain formation, have poor temporal stability and are often very fragile. The next approach is the formation of crystals. Single crystals may provide a high degree of ordering as well as a large connection of chromophores per unit volume, together with a stable structure. But the fabrication of single crystals of good quality is often a very tedious and time consuming procedure.NLO materials find different applications in modern technology. The NLO material with susceptibility $\chi 2$ has application in optical rectification in hybrid bistable device[12], electro-optic (Pockels) effect in modulators, variable phase retarders and frequency doubling in harmonic generation device[13], frequency mixing in parametric amplifiers and IR converters[14], AC electrooptic effect in optical bistability, AC Kerr effect in phase conjugation[15], Frequency tripling in deep UV conversion[16], laser fusion[17] and frequency conversion of ultra short pulses[18]. Marder[19] has reviewed a variety of applications of NLO materials, such as modulation of optical signal facilitated by electro-optic effect (where the refractive index of a material changes in response to an applied electric field), micro-fabrication, sensing, imaging and cancer therapy facilated by multi photon absorption (where molecules simultaneously absorb two or more photons of light), the two photon absorption (TPA) and femto-second laser pulses high density 3D optical memory[20], two photons scanning microscope[21] and demonstrated the 3D micro-modification and tera bit /cm3 3D optical data storage. The nonlinear optical phenomenon of SHG has intrinsic sensitivity to the voltage across a biological membrane. The second order NLO process has been used to monitor membrane voltage by Bouevitch et al.[22].

Ideal NLO crystals are assessed by the set of properties for the fabrication of devices and it may satisfy certain criteria. These are (i) large NLO coefficients (ii) moderate birefringence (iii) small walk-off effect (iv) large angular, spectral and temperature bandwidths (v) wide transparency range for operating.

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