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Development of Nonlinear Optics

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Abstract:

Nonlinear Optics (NLO) is the study of interaction of intense electromagnetic field with materials to produce modified fields that are different from the input field in phase, frequency or amplitude. The goal of nonlinear optics is mainly the investigation of the new phenomena and effects in the interaction process of strong laser and materials, including a deep understanding of the causes and the process regularity and their possible applications in the development of disciplines. In the past two decades, people have made significant progress in nonlinear optical materials. Large nonlinear properties have been used in various photonic and optoelectronic applications such as optical communication, optical information processing, optical data storage, pulsed laser deposition, and optical limiters. This article gives the brief summary about the basic concepts of nonlinear optics and different nonlinear optical phenomenon.

Introduction:

Nonlinear Optics (NLO) is the study of interaction of intense electromagnetic field with materials to produce modified fields that are different from the input field in phase, frequency or amplitude. When the light is not strong, the optical response of a material usually scales linearly with the amplitude of optical electric field. However, at high optical powers, the optical properties of material will be changed more rapidly and are no longer linearly related to the intensity of the incident light. As result, nonlinear optical effects will occur [1]. The goal of nonlinear optics is mainly the investigation of the new phenomena and effects in the interaction process of strong laser and materials, including a deep understanding of the causes and the process regularity and their possible applications in the development of disciplines.

The history of NLO research can be backed to the year 1875 when John Kerr and his co-workers discovered the quadratic electro optic effect in glass (quadratic change in refractive index of material with the application of electric field) [2]. Subsequently, linear electro optical effects were discovered by Pockels in 1893 [3]. As NLO effects are significant only for high intensities (for low intensity field, the system is almost linear for applied stimulus), the first experimental work became possible only after discovery of LASER in 1960 [4]. Peter A Frankel with his co-workers observed second harmonic generation in quartz crystal (doubling of frequency of input beam) [5]. Initially, the progress of NLO was limited to new nonlinear effects and optimal configuration, later with application of NLO and investigation of new materials for NLO and mechanisms of nonlinearity.

Nonlinear optics has a great value and far-reaching scientific significance. In the past two decades, people have made significant progress in nonlinear optical materials. Large nonlinear properties have been used in various photonic and optoelectronic applications such as optical communication, optical information processing, optical data storage, pulsed laser deposition, and optical limiters. Nonlinear optical (NLO) materials are crucial for frequency conversion, and have broadly facilitated the era of optoelectronics [6]. NLO materials find applications in telecommunication [7], spectroscopy [8], and environmental analysis [9]. In the field of spectroscopy, they are capable of high resolution photoemission and lithography [10]. NLO materials have been studied in the past decade for therapeutic applications in eye protection [11] and nuclear or magnetic resonance imaging [12].

Linear and Nonlinear optics:

When a beam with electric field E passes through the medium, it induces polarization (electric dipole moments per unit volume) in the medium. Each constituent molecule acts as a dipole with a dipole moment P_i and the total dipole per unit volume 'P' is given by

$$P = \sum_i P_i \qquad \text{--- (1)}$$

This induced polarization contains lights effect on the medium and the mediums effect back on the light waves. This induced polarization contains linear optical effects and also NLO effects. The effect of external field on the molecular dipoles depends both on the properties of the medium and on the field strength. At low intensity (or low field strength), the induced polarization is proportional to the electric field.

$$P = \varepsilon_0 \chi^{(1)} E \qquad \text{--- (2)}$$

where, $\chi^{(1)}$ is the linear susceptibility that describes linear optical effects.

The relation (2) is valid for field strengths for conventional sources. With sufficiently intense laser radiation, the equation (2) does not hold good and has to be generalized to,

$$P = \varepsilon_0 \left(\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right) \qquad \text{--- (3)}$$

Where, $\chi^{(2)}$ & $\chi^{(3)}$ are frequency dependent second and third order susceptibilities.
Let, the electric field incident on the medium has the form,

$$E = E_0 \cos \omega t \qquad \text{--- (4)}$$

Where, E_0 is the amplitude and ω is oscillating frequency.
Hence we can write equation (3) in the form,

$$P = \frac{1}{2}\varepsilon_0 \chi^{(2)} E_0^2 + \varepsilon_0 \left(\chi^{(1)} + \frac{3}{4}\chi^{(3)} E_0^2 \right) E_0 \cos \omega t + \frac{1}{2}\varepsilon_0 \chi^{(2)} E_0^2 (1 + \cos 2\omega t) + \frac{1}{4}\varepsilon_0 \chi^{(3)} E_0^3 \cos 3\omega t + \cdots \qquad \text{---}$$

(5)

The first term is a constant term. It gives rise to a dc field across the medium, with zero frequency called as optical rectification. This effect is of comparatively little practical importance. The second follows the external polarization and is called first or fundamental harmonic of polarization. The third oscillated at frequency 2ω and is called second harmonic of polarization and the fourth oscillated at frequency 3ω and is called third harmonic of polarization and so on.

Second-order nonlinear optical processes:

The discovery of second-harmonic generation (SHG) in 1961 by Franken *et al.* [5] marked the beginning of the field of nonlinear optics. In 1965, ref. [13] reported the non- linear light scattering in a quartz crystal generating light with frequency twice that of the incident beam. Difference-frequency generation by a KDP crystal using non-collinear light beams was also reported in 1965 in ref. [14]. Apart from second-harmonic generation, the effects that result from second-order nonlinearity or a non-zero $\chi^{(2)}$ include sum- and difference-frequency generation, optical parametric oscillation and spontaneous parametric down conversion. Material symmetry plays a significant role in determining the second-order response as only non-centrosymmetric materials, or materials lacking inversion symmetry show a second-order response.

Second Harmonic Generation:

Second harmonic generation (SHG) is a nonlinear optical process that results in the conversion of an input optical wave into an output wave of twice the input frequency. The light propagated through a crystalline solid, which lacks a center of symmetry, generates light at second and higher harmonics of the applied frequency. Such frequency doubling processes are commonly used to produce green light (532 nm) from, for example, a Nd: YAG laser operating at 1064 nm. This important nonlinear property of non-centro symmetric crystals is called second harmonic generation. The schematic and energy level diagram for SHG is shown in fig (1)

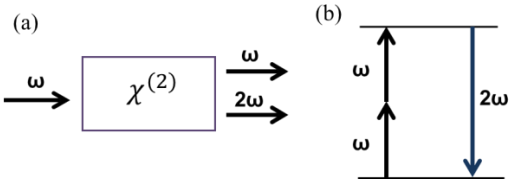

Fig: 1 (a) Schematic and (b) Energy level diagram of SHG process

Phase Matching:

The intense development of research on the mechanism of generation of optical harmonics in crystals and media in which such generation is effectively realizable, has indicated the importance of phase relation between the fundamental and generated harmonics, as they propagate in crystals having optical dispersion (15). It was observed that the efficiency of the generation of the harmonics depends not only on the intensity of the exciting radiation, but also on its direction of propagation in the crystal. The efficiency of all second-order processes depends on the crucial criterion of phase-matching. For frequency doubling the criteria for phase matching is $n_{2\omega} = n_{\omega}$.

Third Harmonic Generation:

The Third harmonic generation **(**THG) is a third-order NLO process that requires three photons (ω) in order to generate one photon at the tripled frequency (3ω). Unlike SHG, THG is not restricted to non-Centro symmetric structures. The schematic and energy level diagram for THG process is shown in fig (2).

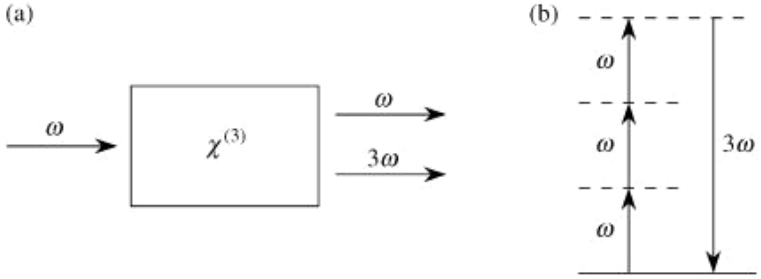

Fig: 2 (a) Schematic and (b) Energy level diagram of THG Process

Nonlinear Index of Refraction

Third order nonlinear polarization contains a term which describes a nonlinear contribution to the polarization at the frequency of the incident field. This term leads to a nonlinear contribution to the refractive index experienced by a wave at frequency ω. It can be defined as a change in the refractive index or the spatial distribution of the refractive index of a medium due to the presence of optical waves.

Many of the interesting phenomena and applications of nonlinear optics emerge on account of nonlinear index of refraction. Several interesting phenomena include:

Optical Kerr effect: The change in index is proportional to the time average of the optical electric field squared i.e. the change in index depends linearly on the optical intensity [1].

Nonlocal effect: The index change depends on the time averaged field, or optical intensity, but is due to a gradient in the field (e.g. thermal gradient) or long range order in the medium (e.g. density variation in case of liquid) [1].

Saturation: The change in index is caused due to absorption of optical energy by the medium [1].

Several interesting applications include: Nonlinear spectroscopy, Optical switching, Optical logic gates, Optical communications, Optical limiting, passive laser mode-locking and Waveguide switches, modulators, etc. The mechanisms responsible for nonlinear index of refraction are electronic polarization, Raman induced Kerr effect, molecular orientational effects, electrostriction, thermal effects etc.

Nonlinear absorption:

Nonlinear absorption refers to the change in transmittance of a material as a function of intensity or fluence. At sufficiently high intensities, the probability of a material absorbing more than one photon before relaxing to the ground state can be greatly enhanced. In addition, population redistribution induced by intense laser fields leads to interesting counter plays of stimulated emission and absorption, complicated energy transitions in complex molecular systems and the generation of free carriers in solids. These phenomena are manifested optically in a reduced (saturable) or increased (reverse saturable) absorption.

Two Photon Absorption:

Two-photon absorption (TPA) takes place when the system simultaneously absorbs two photons in order to make transition from the ground state of a system to a higher-lying state. The two photons may be from the same exciting field or may be from two different exciting fields (with two frequency fields). This

process involves different selection rules than those of single-photon absorption. Hence TPA spectroscopy provides great avenues in studying the excited states of systems. Although the transition does not involve a real intermediate state, often there are impurities present that will produce a small amount of linear absorption. It should be understood that this absorption does not contribute to the transition to the final state of the process but only serves as an additional loss mechanism. The energy level diagram for two photon absorption is shown in fig (3)

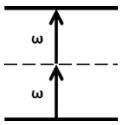

Fig [3]: Energy level diagram for two photon absorption

Excited State Absorption

When the incident intensity is well above the saturation intensity, then the excited state can become significantly populated. In systems such as polyatomic molecules and semiconductors, there is a high density of states near the state involved in the excitation. The excited electron can rapidly make a transition to one of these states before it eventually transitions back to the ground state. There are also a number of higher-lying states that may be radiatively coupled to these intermediate states, and for which the energy differences are in near-resonance with the incident photon energy. Therefore, before the electron completely relaxes to the ground state, it may experience absorption that promotes it to a higher-lying state. This process is called excited state absorption. It is observable when the incident intensity is sufficient to deplete the ground state significantly.

Saturable Absorption

When the absorption cross-section of the excited state is smaller than that of the ground state, the transmission of the system will be increased when the system is highly excited. This process is called saturable absorption (SA). Saturable absorption involves a decrease in absorption at high optical intensities and it is usually observed in materials that are strongly absorbing at low light intensities. Saturable absorption occurs when the upper state of the absorbing transition gains enough population to become filled, preventing the transfer of any more population into it. Saturable absorption is used to mode lock solid state and pulsed dye lasers. It can also be used with four wave- mixing interactions to produce optical phase conjugation.

Reverse Saturable Absorption:

When the absorption cross-section of the excited state is larger than that of the ground state, then the system will be less transmissive when excited. This gives the opposite result as saturable absorption and is thus called reverse saturable absorption (RSA).

Free Carrier Absorption

When the absorption cross-section of the excited state is larger than that of the ground state, then the system will be less transmissive when excited. This gives the opposite result as saturable absorption and is thus called reverse saturable absorption (RSA).

Optical limiting:

Optical limiters are devices that transmit light at low input fluences or intensities, but become opaque at high inputs. Optical limiters have applications in optical pulse shaping, pulse compression and protection of human eye and optical sensors from intense laser pulses. One of the desirable attributes of an optical limiter is low threshold. An ideal optical limiter will have a linear transmission upto a threshold input fluence I_{th}, which varies for different materials.

The important processes causing optical limiting effects are reverse saturable absorption, two photon absorption, free carrier absorption, nonlinear refraction, photo refraction and induced scattering. TPA and TPA induced FCA has been extensively studied for optical limiting. Thresholds of TPA materials depend on β and are generally much higher than those observed in RSA materials. However TPA materials do not saturate as RSA materials.

Nonlinear refraction (NLR):

Nonlinear index of refraction is the change in refractive index or the spatial distribution of the refractive index of a medium due to the presence of optical waves and has generated significant and technological interest. It has been utilized for a variety of applications such as nonlinear spectroscopy, correcting optical distortions, optical switching, optical logic gates, optical data processing, optical communications, optical limiting, passive laser mode-locking, wave guide switches and modulators. Several physical mechanisms that contribute to the NLR include electronic polarization; Raman induced kerr effect, molecular orientational effects, electrostriction, population redistribution, thermal contributions, cascaded second order effects and photorefractive effect.

Conclusion:

To meet the requirements for integrated nonlinear optics, different NLO materials are being developed. Second order NLO materials are used in optical switching (modulation), frequency conversion (SHG, wave mixing), and electro-optic applications, especially in EO modulators. Metal nanocomposites and semiconductor nanocomposites in which metal or semiconducting nanoparticles are dispersed in ceramic or polymer matrix are also showing outstanding results especially for third order nonlinearity which are useful for various applications. NLO thin film materials are also prompting for new technologies. The breadth and depth of NLO is truly remarkable and due to this fact, instead of attaining saturation, NLO now spans from macro system to nano system and is now an interdisciplinary field of research.

References:

1. R.W. Boyd; Nonlinear Optics (Academic press, 2002).
2. J. Kerr, Phil. Mag. 50, s.337-348 i s. 446-458 (1875).
3. G. New, Introduction to Nonlinear Optics, Cambridge University Press (2011).
4. T. H. Maiman, Nature 187, s. 493-494, (1960).
5. P. A. Franken, A. E. Hill, C. W. Peters, G. Weinreich, Phys. Rev. Lett. 7, 118-119, (1961).
6. L.R. Dalton, et al., Organic Electro-Optics and Photonics: Molecules, Polymers and Crystals, Cambridge University Press, (2015).
7. G. Zerbi, Organic Materials for Photonics: Science and Technology, Elsevier, (2013).
8. J. Yu, et al., ACS Appl. Mater. Interfaces 12 (40), 45023 - 45035 (2020).
9. B. Zhang, et al., Chem. Mater. 32 (15) 6772 – 6779 (2020).
10. B. Zhang, et al., Angew. Chem. 129 (14), 3974 – 3977 (2017).
11. C. Zheng, et al., Appl. Phys. B 124 (1) 17(2018).
12. Y. Qiang, et al., J. Nanoparticle Res. 8 (3) (2006) 489-496.
13. Terhune R. W., Maker P. D. and Savage C. M., Phys. Rev. Lett., 14, 681 (1965).
14. Van Tran N., Spalter J., Hanus J., Ernest J. and Kehl D., Phys. Lett., 19, 4 (1965).
15. Franken, P. A. and Ward J. F. Rev. Mod. Phy. 35, 23 (1963)