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An Analysis of an Application of Quantum Dots in Non Linear Optics

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ABSTRACT:

Remarkable effort and progress have been made in semiconductor quantum dots and their applications [1-21]. Size confinement results in many basic properties of substance which has been utilized in designing better products. Bulk materials have constant physical properties regardless of its size, while in nano particles the size often dictates the physical and chemical properties. Because of faster response speed, nano particles are proved to be very efficient candidates for many opto-electronic phenomena such as nano LED. In this context, it is interesting to think whether non- linear optical techniques can offer similar perspectives. With the advent of Laser beams, practically a large number of interesting non-linear optical phenomena could have been possible to be investigated[1]. One such important non-linear optical effect is Second Harmonic Generation (SHG) which is second order non-linear phenomena. The generation of the Second Harmonic is nothing but the frequency doubling. If, say,red light from a ruby laser (694.3nm) is allowed to go through a KDP crystal, a part of it gets converted into a new light wave of wavelength 347.14nm (ultraviolet) ie half the wavelength of the incident wave.

KEYWORD: Linear Nonlinear Optics, quantum confinement. Literature, KDP crystal

INTRODUCTION:

In the present work, an attempt has been made to test the application of our quantum dots in area of nonlinear optics. In non linear optics, the study has been confined only to the application of our quantum dots for Second Harmonic Generation although a number of other applications of quantum dots are possible in non linear optics. In fact, several non-linear applications have been demonstrated to benefit from the size confinement features of quantum dots1. Numerous facets of optical non-linearity in nano crystals in relation to the impacts of quantum confinement have recently been studied using optical techniques. Second order optical non-linearities of nanostructures have gotten substantially less attention, according to a literature review10. Practically a huge number of intriguing non-linear optical phenomena may have been studied with the development of laser beams1. Second Harmonic Generation (SHG), a second order non-linear phenomenon, is one such significant non-linear optical effect.

LINEAR AND NON-LINEAR OPTICS:

Light wave is an electromagnetic wave and when it propagates through an optical medium, the oscillating electric field **E** exerts a polarizing force on the electrons of the atoms constituting the medium. The electric field induces electric dipoles in the medium, the dipoles orienting them along the field direction and the dielectric medium is said to be polarized. The optical response of a medium is expressed in terms of induced polarization **P**. For a linear medium the relation between the induced polarization **P** and the electric field **E** of the incident radiation is linear:

$\mathbf{P}=\varepsilon_0 \chi^{(1)}\mathbf{E}$ where $\chi^{(1)}$ is the linear susceptibility and ε_0 is the permittivity of free space [2,9].

With ordinary source of light, the electric field strength cannot affect theoptical parameters of the medium as the ratio **P/E** remains independent of intensity of incident light. **P** is proportional to **E** and a plot of **P** vs **E** is a straight line (linear). This is illustrated by dotted line. (Figue-5.1)

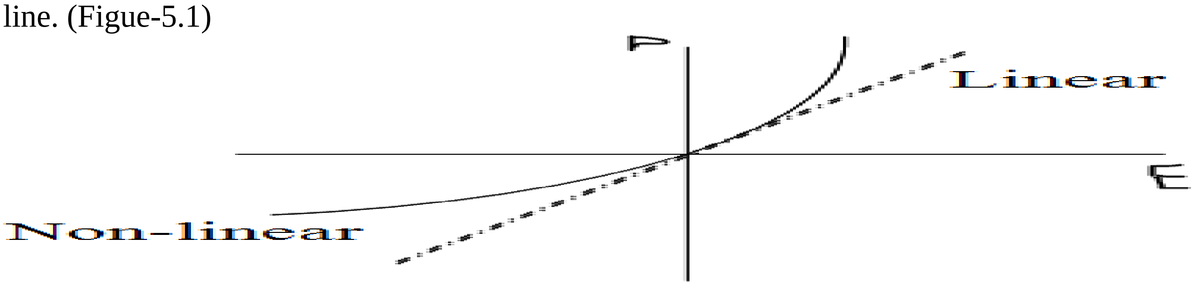


Figure 5.1 Illustration of linear & non-linear relation between P&E

The medium that obeys the above principles are known as linear medium and their optical behavior is known as linear optics. In this case, properties such as susceptibility (χ), refractive index (n) etc. of the medium are independent of the field intensity **E**. The situation however drastically changes if the incident light becomes highly intense. Polarization then is no longer proportional to the electric field but becomes saturated and acts non-linearly. This non linearity of **P** with **E** occurs with high field strengths of light source like that of laser light. The non linearity of **P** affects the optical parameters of the medium when χ, η, $\boldsymbol{\varepsilon_0}$ etc. are no longer constants, but becomes functions of the field strength **E** and under these circumstances the optical behavior of the medium is known as non linear optics. This is illustrated by curved line in figure 5.1

In nonlinear optics, the response of the medium is described as a Taylor expansion of the polarization **P** in powers of the electric field **E**. Using the Einstein summation convention, each component P_k (k = x, y, z) of the polarization can be expressed as:

$$P_k = \varepsilon 0 \left(\chi^{(1)}_{ik} E_i + \chi^{(2)}_{ijk} E_i E_j + \ldots\ldots\ldots\ldots \right.$$

Here, the coefficients $\chi^{(n)}$ correspond to the tensor of the n-th order nonlinear process. For most practical applications let consider a specific non-linear process in a well defined direction, the tensor can be reduced to a single effective nonlinear coefficient[2].

Let us consider only the second order term and calculate the resulting nonlinearpolarization, i.e. $P_k(NL) = \chi^{(2)}_{ijk} E_i E_j$ for a input field at frequency ω. Any medium, in fact, becomes non-linear if the field of incident radiation is very high. Linear optics is only a first order approximation of non-linear optics. The development of the laser provided intense source of monochromatic and coherent light that stimulated extensive research and applications in the field of nonlinear optics.

Second Harmonic Generation:

In Second Harmonic Generation photons (light) interact with a non-linearmaterial (medium) effectively to combine to form new photons with twice the energy of incident photons and thus twice the frequency and half the wavelength of the initial photons. Second Harmonic Generation proceeds in two steps; the input wave generatesa set of dipoles (inside the medium /material) that radiates weak light signal of twice the frequency of the input light signal. As the input light signal (optical signal) has well defined phase and amplitude at every point inside the material at a given time, relative phase of the induced dipoles is fixed. To obtain the Second Harmonic output from any material (or medium), it is important that the induced dipoles radiate in same phase.

Incident light
(Laser)

Non linear optical material (NLO)

Figure 5.2 Second harmonic generation in NLO.

The Second Harmonic Generation is achieved only when there is a phase matching between pump wave (input excitation) and output signal i.e generated wave. For phase matching, material is to be non-symmetric. In practice II-V1 semiconductorsare centro-symmetric in nature. However, when bulk II-VI semiconductors e.g SnO_2, ZnS and CdS are converted in to quantum dots these are transformed into non- centro symmetric specimen. This is due to the fact that with dimension of only couple of nanometers, the increase of surface to volume ratio results in the breaking of the centrosymmetry at the surface of the material to convert it in to a non centrosymmetric one[5]. This phenomenon causes quadratic hyperpolarisability[5] which is the key reason for the Second order non-linearity through phase matching when maximum transfer of energy from fundamental (Pump) wave to Second Harmonic takes place.

5.2.1.1 Second Harmonic Generation: A mathematical Approach

If a high intense beam of light strikes a medium or material, the later responds in a nonlinear manner and higher optical harmonic is generated in addition to linear optical response. The strongest measurable non-linear optical response is the Second Harmonic Generation (SHG). The generated higher harmonics are functions of the specimen atomic structure.

Condition for Second Harmonic Generation:

The condition for second harmonic generation has been discussed in literature[2].One such approach has been made by loud[3] which explains the second harmonic generation by quantum dots[11.12.13]. However, a more elaborate discussion is available in the literature by Ghatak and Tyegarajon[2]. Hence, to understand the second harmonicphenomenon clearly, the mathematical approach is made according to Ghatak and Tyegarajon[2] in the thesis as follows:

When the electric field of light wave becomes strong then the polarization is no more proportional to electric field, it has also higher order terms.

The Second term is responsible for 2nd order nonlinearity.In scalar form.

$$P = \epsilon_0 \chi E + \epsilon_0 \chi^{(2)} E^2 + \text{-----------} \quad (1)$$

$$E = E_1 \cos(kz - \omega t) \text{------------} \quad (2)$$

Equation (2) initiates the di-pole polarization effects. Polarization is nothing but di-pole moment/volume which implies that dipoles are oscillating at frequency 2ω and it generates electromagnetic wave at 2ω frequency. This is the origin of 2ω.

Incident wave $\omega \longrightarrow$ Nonlinear medium $\longrightarrow 2\omega$

Thus, the total electric field inside the material (medium) is generating andgenerated and is given as

$$E = E_1 \cos(k_1 z - \omega t) + E_2 \cos(k_2 z - 2\omega t) \text{.......} \quad (3)$$

$k_1 = \frac{\omega}{c} n(\omega) = \frac{\omega}{c} n_1, \; n_1 \rightarrow$ Refractive index of the medium (air)

$k_1 \rightarrow$ Propagation constant of the medium at frequency 2ω given by

$k_2 = \frac{2\omega}{c} n(2\omega) = \frac{2\omega}{c} n_2, \; n_2 \rightarrow$ Refractive index of the non-linear medium.

It is important to note that refractive index of the medium (material) dependson frequency.hen we launch frequency ω inside the medium, we find, throughE^2 term electric field at 2ω also generated. For second harmonic generation one must satisfy some condition called phase matching condition. If this is not satisfied the efficiency of generation of new frequency will be very little. This is one of the essential and necessary criterion for Second Harmonic Generation. Let explain the meaning of phase matching condition mathematically and physically. For non-linear media.

$\omega \longrightarrow \boxed{\chi^{(2)} \neq 0} \longrightarrow 2\omega$

For a medium (material) with centre of inversion symmetry $\chi^{(2)} = 0$, for medium without centre of inversion symmetry, $\chi^{(2)} \neq 0$ and hence can generate 2nd order non-linearity i.e 2ω. Actually the asymmetry of the molecular structure is the primary source of second order non-linearity. This is the second essential requirement for Second Harmonic Generation.

Block Diagram of Experimental Setup for Observation of Second Harmonic Generation by Quantum Dots:

Figure 5.3 represents the block diagram of experimental setup to observe second harmonic generation by quantum dots. The experiment is performed in air medium at room temperature. To carry out the study, the quantum dot film of area (1cm x 1cm) is cut, made fixed with sample holder and illuminated by input pump wave generated by a xenon lamp. The lamp emits light and the monochromator selects the specific desired wavelength of input pump wave (light). Further, it has been observed in the present study that with input pump wave of wavelength lower than 700 nm does not cause secondharmonic generation in our quantum dots rather it produces photoluminescence. It is to be noted that the pump wave from 200 to 545 nm causes photo luminescence at around 585 nm and 580 nm in case of SnO_2 and ZnS quantum dots respectively whilethe input pump wave from 200 nm to 630 nm produces luminescence at around 680 nm in CdS quantum dots as discussed in chapter 4. The experiment is performed with the help of a fluorescence spectrophotometer (Make: Perkin Elmer, model: LS 45)

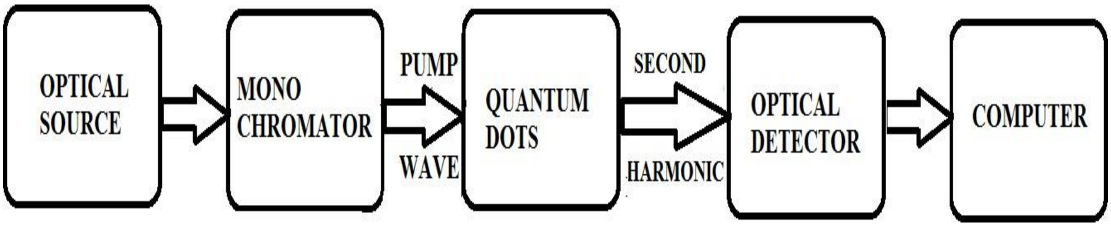
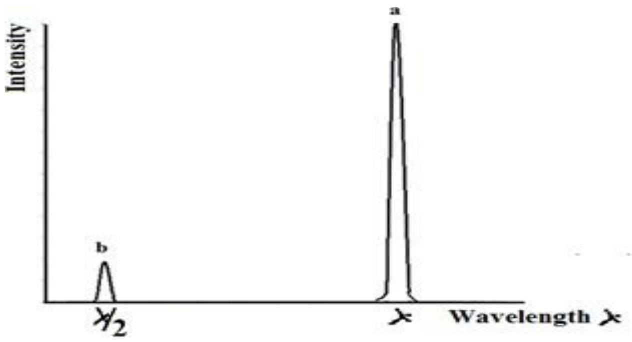


Figure 5.3 Block diagram of experimental setup for second harmonic generation with



quantum dots**Figure 5.4** Pattern of spectrum containing input and input signal

In figure 5.4, „a" indicates the input pump wave while "b" indicates the output wave. If the output wave be the second harmonic of input then output "b" will occur at half the wavelength of input signal "a". Thus, if the wavelength of input signal be λ, the wavelength of output signal will be λ/2.

CONCLUSION:

The quantum dots namely SnO_2, ZnS and CdS find their applications as secondharmonic generators which is one the potential applications of quantum dots in the field of non linear optics. The input excitation (pump wave) ranges from 700 nm to900 nm with intensity from 0-210 (a.u). However, SnO_2/SBR quantum dots do not produce second harmonic in response to pump wave in the said range. Furthermore, it has been observed that the intensity of second harmonic is highest in case of CdS and lowest in SnO_2 quantum dots. The phase matching condition of quantum dots isexamined with respect to air and hence the application is tested in the air medium only.

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