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RESEARCH ARTICLE

SECOND HARMONIC GENERATION IN CdS NANO QUANTUM DOTS PREPARED BY CHEMICAL METHOD

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ABSTRACT

Quantum confined semiconductor structures have attracted recent attention of researchers as potential candidate for nonlinear optical applications. There has been a rapid research activity in the field of nonlinear optics. One such nonlinear effect is Harmonic Generation. In this paper, we report the generation of second harmonic by CdS semiconductor quantum dot prepared by simple chemical route. SHG is one of the important 2nd order non-linear optical effect. Due to size confinement, high quantum efficiency gives high intensity beam at half of the wavelength range on exposing the quantum dots in UV/VIS region of optical wavelength. The observed Second Harmonic Generation is achieved only when there is a phase matching between pump wave and generated wave.

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INTRODUCTION

The optical properties of quantum confined semiconductor structure such as quantum dots have gathered much interest since the first experimental results of Ekimov and Onuschenko (Gary Wiederrecht, 2010; Butcher and Cotter, 1990; Franken *et al*., 1961; Byung-hee choi *et al*., 2007; Dey *et al*., 2010; Michal Jacobsohn and Uri Banin, 2000; Michal Jacobsohn and Uri Banin, 2000; Andrew R. Barron; Nath *et al*., 2009; Dey *et al*., 2010; Mohanta *et al*., 2003; Chattopadhyay and Banerjee, 2009). The enormous potential of semiconductor quantum dots as non-linear optical materials was first pointed out by Jain and Lind. With the advent of Laser beams, a large number of interesting non-linear optical phenomenon could have been possible to be investigated (Gary Wiederrecht, 2010; Butcher and Cotter, 1990; Franken *et al*., 1961; Byung-hee choi *et al*., 2007; Dey *et al*., 2010; Michal Jacobsohn and Uri Banin, 2000; Michal Jacobsohn and Uri Banin, 2000; Andrew R. Barron; Nath *et al*., 2009; Dey *et al*., 2010; Mohanta *et al*., 2003; Chattopadhyay and Banerjee, 2009). One such important non-linear effect is Second Harmonic Generation (SHG). The generation of second harmonic is nothing but the Frequency Doubling. If ,say, red light from ruby laser (694.3nm) is allowed to go through a KDP crystal, apart of it gets converted into a new light wave of wavelength 347.15 nm (ultraviolet) ie half the wave length of the incident wave. This was first demonstrated in 1961 by Franken and co-workers. The experiment of SHG (Butcher and Cotter, 1990; Franken *et al*., 1961; Byung-hee choi *et al*., 2007; Dey *et al*., 2010; Michal Jacobsohn and Uri Banin, 2000) marked the birth of non-linear optical study. In the phenomena a coherent input generates a coherent output of double the frequency of input one. Literature reports the generation of Second Harmonic by metal quantum dots which act as non-linear optical material (Butcher and Cotter, 1990). Quantum dots are basically three dimensionally confined system with sizes up to 30nm. In recent years, optical techniques have been used to investigate the many aspects of optical non-linearity in nono crystals in relation with the effects of quantum confinement such as linear and third order nonlinerities. But Second Order optical non-linearities of the nano crystals have received much less attention (Andrew R. Barron). Considering that field of study of SHG by semiconductor quantum dots may be an interesting area of research activity, we have taken this task for our investigation. In semiconductor quantum dots due to quantum confinement, it has been observed specifically i) an increase in band gap ii) an increase in S/V iii)luminance property. Size dependence of Second Harmonic Generation in CdSe nano crystal quantum dots has been studied by Michel Jcobsohn and Uri Banin (2000). Here in this paper it is an attempt to study the effect of very popular Binary semiconductor Compound CdS (II-VI group) quantum dots prepared by chemical method for Second Harmonic Generation on illuminating the specimen with intense monochromatic Laser source in the UV/VIS range and detecting the optical output at half wavelength of that of input.

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Theory

A light wave is an electromagnetic wave and when it propagates through an optical medium oscillating electric field E gives rise to polarization of the medium. The non-linear polarization response of a molecule can be expanded as a power series of the inducing optical field of strength E as

$$P = \epsilon_0 (\chi_1 E + \chi_2 E^2 + \chi_3 E^3 + \ldots\ldots) \quad \text{----------(1)}$$

Where χ_n is the nth order susceptibility. The order of magnitude of susceptibility drops rapidly with n. The lowest order term (n =1) gives the linear dielectric function. The second term ($\epsilon_0 \chi_2 E^2$) is responsible for second harmonic generation.

Synthesis of CdS quantum dots on PVA Matrix

5 grams PVA are dissolved into 100 ml double distilled (D/D) water. The mixture is taken in a three necked flask fitted with thermometer pocket and N_2 inlet. The solution is stirred in a magnetic stirrer at a stirring rate of 200 rpm in the constant temperature of 70^0 C for 3 hours. Thus, a transparent water solution of PVA has been prepared. The solution is degassed by boiling N_2 for 3 to 4 hours. Similarly, $CdCl_2$ solution is made by dissolving 5 gms of $CdCl_2$ in 100 ml D/D water. Next PVA solution and $CdCl_2$ solution are mixed in the volume ratio of 2 :1 and few drops of HNO_3 is added to the mixture and stirred at the rate of 250 rpm at a constant temperature of 55^0C while 2Wt % aqueous solution of Na_2S is put into it by dropping funnel slowly unless the whole solution turns into yellow colour. The solution is kept in dark chamber at room temperature for 12 hours for its stabilization followed by its casting over glass substrate and drying in oven at 50^0 C. This film contains CdS quantum dot[6] embedded in PVA matrix.

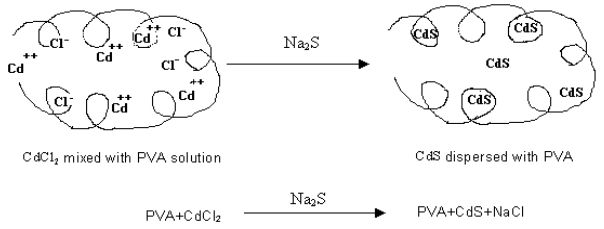


Fig.1.

Optical absorbance spectroscopy

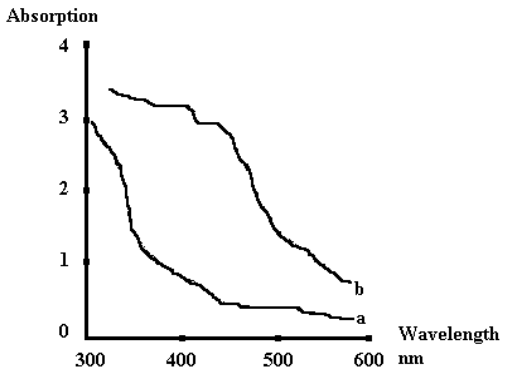


Figure 2. Optical spectra of CdS. a: quantum dot in PVA, b: bulk

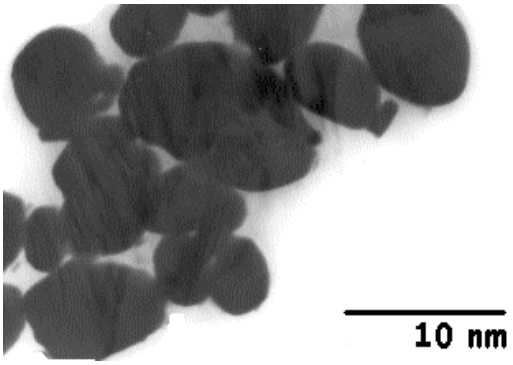
RESULTS AND DISCUSSION

Optical absorption spectroscopy (using Perkin Elmer Lambda 35) display strong blue shift (Figure -2) in the absorption edge at 370 nm which indicates the formation of nanostructure. The appropriate size can be assessed by using the following hyperbolic band model (Dey *et al*., 2010). The model yeilds the average quantum dot size at 8 nm. Formula for hyparabolic Band Model is

$$R = \sqrt{\frac{2\pi^2 h^2 E_{gb}}{m^*(E_{gn}^2 - E_{gb}^2)}}$$

where Egb = bulk band gap = 1242/λgb, λgb= bulk transition wavelength, Egn=QD b and gap=1242/λgn, λgn=wavelength corresponding to the strong absorption edge of the quantum dots and m* effective mass of the quantum dots and R is the radius of quantum dot, High resolution Transmission Electron Microscopy (HRTEM) (using JEOL, 100CXII, 100Kv) shows the particle size of the sample (Figure-4). Average diameter of the particles are found around 9 nm.

Transmission Electron Microscopy (TEM)



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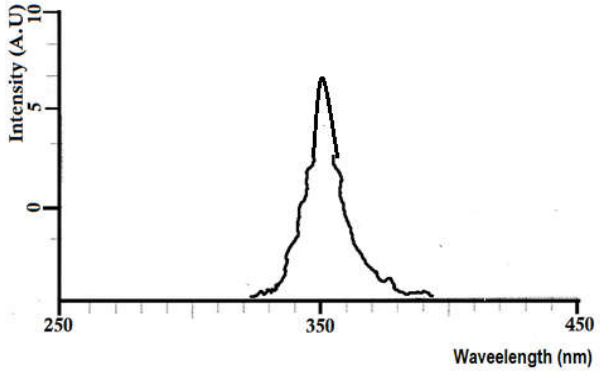

Figure 3.

We believe this to be second harmonic generation because of the following facts:

(1) The asymmetry of the molecular structure is the primary source of the second order optical nonlinearity

and second harmonic can be generated by non-linear optical material with non-centro symmetric molecular organization (Michal Jacobsohn and Uri Banin, 2000; Gary Wiederrecht, 2010).

(2) The efficiency of nano objects to convert fundamental frequency photons (pump wave) into second harmonic photons is determined by the quadratic hyperpolasability $(\beta)^1$ as clear in the equation (Gary Wiederrecht, 2010)

ZnS is a non centrosymmetrical material even in bulk from and hence is a right candidate to generate second harmonic. Normally the hyperpolasability (β) of ZnS lies between 10^{-25} to 10^{-26} esu. When the size is reduced specially within 10 nm the value of β increases remarkably and goes as high as This is because the surface properties of the particles confined within 10 nm are observed through the surface contribution of β [1]. Also, the optical output intensity of Cds quantum dots quadritically depends on the input incident optical field as in fig 4. In Photoluminescence study of the sample, obtained outputs against different input intensities show excellent agreement to quadratic fit (Michal Jacobsohn and Uri Banin, 2000).

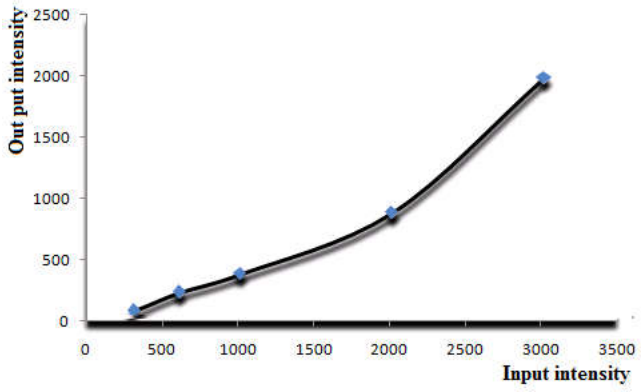

Figure 4.

Conclusion

ZnS quantum dots fabricated by chemical method is within 10 nm of particle size range show high second order hyperpolasability resulting in generation of Second Harmonic wave.

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