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RESEARCH ARTICLE

THEORETICAL APPROACH TO SUBSTANTIATE THE GENERATION OF SECOND HARMONIC FREQUENCY BY CdS QUANTUM DOTS

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ABSTRACT

Cadmium Sulphide (CdS), a II-VI group binary compound is prepared in laboratory through chemical method. Quantum dot sizes are restricted with uniform size distribution and the synthesis is done along with poly vinyl Alcohol (PVA) Matrix. Absorption Spectroscopy in the UV range, XRD and HRTEM studies are carried ooutto characterise its nano properties : size and concentration. The second order non-linear effect expected to be produced by the sample leads to its application as Second Harmonic Generation. The sample depicted successfully in photoluminescence study. Also, in this paper an attempt has been made to justify the generation of second harmonic generation in our as-prepared CdS quantum dots.

INTRODUCTION

Invention of laser Light by in 1960 quickly shifted physicist interest for research in the non-linear fields (Franken *et al.,* 1961). In pre- Laser era, light wave with a low intensity is not able to affect atomic fields to the extent of changing optical parameter. But high degree of coherence of laser radiation ceases the linear relationship between electric polarisation P and the field strength E. New features are observed and eventually leads to new applications. And many works with metal nano-particles in the field of electronics were already reported. Research work on photoluminescence[5], electroluminescence (Nath *et al.,* 2009) study with semiconductor nano particles were carried out by many research scholars After literature survey, we have found that work on non-linear effect such as frequency mixing, frequency doubling etc (Gary Wiederrecht, 2010; Andrew). Were reported to be done with some compound nano particles both metal and semi conductor by some groups (Tiwari, 2006). But many aspect of them were yet to be remained unexplored for non-linear effect study. CdS is considered again for second harmonic generation effect and to establish phase matching condition found from the experimental result.

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It has been experimentally observed that CdS can act as a second harmonic generator around a particular wave length in the optical spectrum. The experimental result is well supported by theoretical one also.

Brief theory

At high values of electric field many material show non-linear relation of polarizability. This can be explained by a non-linear susceptibility. These susceptibilities are important in non-linear optics and lead to effects such as Second Harmonic Generation. Taylor expansion of the polarization reaction to electric field which involves non-linear susceptibilities

$$p = \epsilon_0\chi^{(1)}E+\epsilon_0\chi^{(2)}E^2+\epsilon_0\chi^{(3)}E^3+\text{- - - - -}$$

The first susceptibility term $\chi^{(1)}$ corresponds to linear susceptibility. The co-efficient $\chi^{(2)}$, $\chi^{(3)}$ ---- define the degree of non-linearity and are known as non-linear susceptibilities. Suppose the incident wave is of frequency ω_1 Thus inside material, generated S.H wave at frequency $2\omega_1$ radiates an electromagnetic wave of the same frequency which propagates with same velocity as that of the incident wave. The produced wave has the same characteristics of directionality and monochromacity as the incident wave and is emitted in the same direction7. Non-linear susceptibility $\chi^{(2)}$ depends on the

direction of propagation, polarisation of the electric field and the orientation of the optic axis of the crystal. Hence χ must be treated as tensors &P^{NL} therefore may be represented by

$$P^{NL} = \epsilon_0 \sum_{ij} \chi^2_{ijk} E_j E_k$$

Where i, j,k represents the co-ordinates x,y,z

In isotropic crystal χ is independent of direction, hence is a constant[7].Which means $Pi^{NL} = 0$ and hence $\chi^2_{ijk} = 0$

Second Harmonic Generation, therefore, cannot occur in an isotropic medium, nor in centro symmetric crystals. It has been found that only crystals that lack inversion symmetry exhibit SHG.In case of non-centro symmetric materials second order term dominates, other higher order terms may be ignored and for such material, one can write

$$P^{NL} = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2$$

It was observed that the efficiency of generation of harmonics depends not only on the intensity of excitation radiation but also on its direction of propagation in crystals. To generate $2\omega_1$, frequency from ω_1, system must satisfy some condition called phase matching condition

If generated wave propagates a length L in the material, The expression for intensity

$$I \propto \frac{sin^2 \frac{2k_1 - k_2}{2} L}{\frac{2k_1 - k_2}{2}}$$

is sharply peaked about

$$\frac{2k_1 - k_2}{2} = 0$$

i,e when $k_2=2k_1$ or $k_2-2k_1=0$

Where K_1- propagation wave number of incident wave
K_2- propagation wave number of SHG wave

For efficient frequency doubling this relation must be satisfied. This is a phase matching condition

Since $k_2 = \frac{2\omega\, n_{2\omega 1}}{C}$ & $k_1 = \frac{\omega n_{\omega 1}}{C}$

Relation reduces to

$n_{2\omega}=n_{\omega}$

Hence this refractive index criterion is also treated as phase matching requirement (Laud). Now, in general, the threshold of photon absorption of a semiconductor determined by the energy gap where as refractive index is a measure of transparency to the incident photon. Moreover, electronic properties such as atomic polarizability and dielectric constant depend on the refractive index of the materials which ultimately can be calculated from the knowledge of the energy gap

We use the relation between refractive index and energy gap developed by Rabindra as

n =4.084-0.62Eg

Experiment

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.

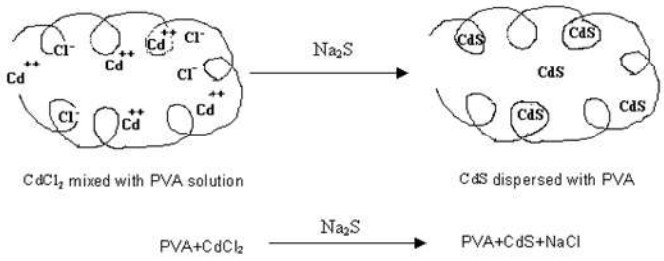

Fig. 1.

RESULTS AND DISCUSSION

Optical absorbance spectroscopy

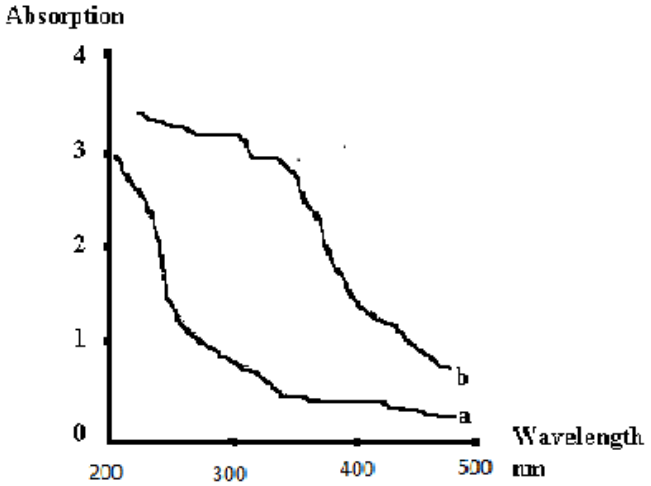

Figure 2. Optical spectra of CdS. a: quantum dot in PVA, b: bulk

Optical absorption spectroscopy (using Perkin Elmer Lamda 35) display strong blue shift (Figure 2) in the absorption edge at 270 nm which indicates the formation of nanostructure. The appropriate size can be assessed by using the following hyperbolic band model [10,16]. The model yeilds the average

quantum dot size at 9 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, band 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,

Transmission Electron Microscopy (TEM)

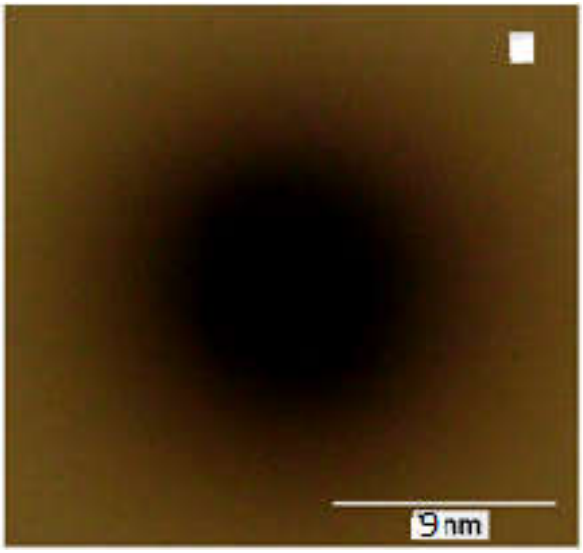

Fig. 3.

High resolution Transmission Electron Microscopy (HRTEM) (using JEOL, 100CXII, 100Kv) shows the particle size of the sample (Figure 3). Average diameter of the particles are found around 9 nm.

X-ray diffraction study

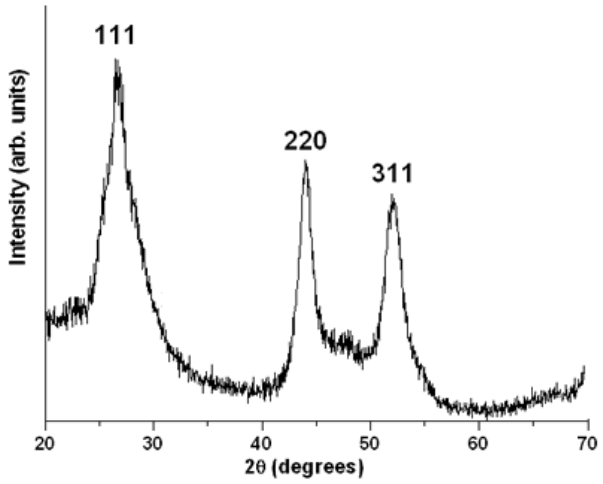

Fig. 4.

Application

To test the second harmonic wave in our prepared CdS quantum dots, the sample is illuminated with monochromatic optical pump wave and the optical output is detected with a photomultiplier tube (PMT) having maximum sensitivity in the optical range from 300 nm to 500 nm. In the present study CdS quantum dots are illuminated with an optical signal (pump wave) of 750 nm and the optical output is detected at an wave length of around 380 nm. The output spectrum is shown in figure-5 below.

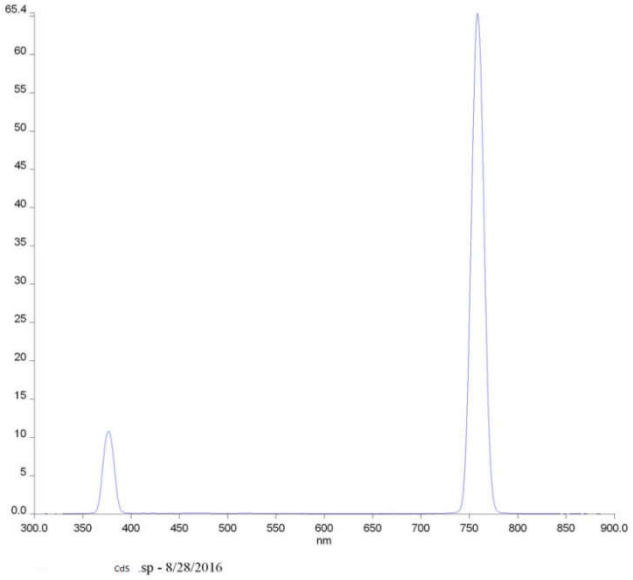

Fig. 5.

Calculation from the relation

n = 4.084-0.62 Eg
= 4.084-0.62x4.6 =1.232

Now for air n_ω =1 and $n_{2\omega}$ =1.232 which follows phase matching condition as desired by theory. Discrepancy in value of refractive index of non-linear material $n_{2\omega}$ is because of dispersion.

Conclusion

CdS quantum dots are prepared though chemical route. Different characterization tools suggest that the prepared quantum odts are of uniform size and are within 10 nm. The as-prepared CdS quantum dots show second harmonic when excited with a pump wave at around 750 nm.

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