

Effect of Particle Size Distribution on Spectral Characteristics of Different Quantum Dots

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Abstract

The impressive progress in the fabrication of low dimensional semiconductor structures during the last decade made it possible to reduce the effective dimensions from three dimensional bulk materials, to quasi two dimensional quantum well, to quasi one dimensional wires, and even to quasi-zero dimensional quantum dots. The quantum confinement effects in such semiconductor systems with reduced space dimension have attracted considerable attention; this is because of the potential application of the dots in electronic and optoelectronic devices. These nanostructures confine carriers in all three dimensions. As a consequence of this, several important characteristics emerge: these include atom-like density of states, large exciton binding energies, enhanced luminescence and oscillator strengths.

Keywords: inhomogeneous broadening, size-distribution, dispersion, line shape function.

INTRODUCTION

When Quantum dots of different sizes exist in a material, de-phasing occurs within a quantum dot homogenous and inhomogeneous broadening is observed in PL spectra due to uniform and non-uniform distribution of quantum dots within the samples [1-6]. The quantum dots (QDs) have been used as an important quantum structure in opto-electronic materials [7-13] due to atomic-like quantum confined electronic energy levels in quantum dots. These studies demonstrate that both the emission and detection devices can be improved by using QD structures. The size distribution property has shown its strong effect on the devices performance and the line shape of photoluminescence spectrum (PL), and has been studied in the past few years[14-19]. In optoelectronic devices, QDs with many layers and high density are grown to improve the quantum efficiency of devices.[20-21] However, it is of great difficulty to obtain QDs with uniform size distribution.[22-25] The most mature method to characterize the size distribution of QDs is to measure the size of every QD on the surface by the atomic force microscopy (AFM) directly. Some observations have shown that the broadening of photoluminescence is resulted from the size distribution of QDs. It is interesting to investigate the quantitative correlation of PL line shape with the size distribution, since this can provide a more precise model for QDs material in device performance design. Generally, a Gaussian size distribution

function is used to derive information about the quantum dot sample under size distribution [1]. It is also evident that as the quantum confinement increases with decreasing particle size, the quantum effect become more pronounced for particle distributions with a smaller mean particle diameter when the geometric standard deviation is kept constant, the particle in the distribution which contribute to absorption are substantially larger than the mean particle diameter, thus reducing the observed band-edge energy[26]. In addition PL emission from a single particle is expected to be narrow, the measured spectra for size distribution exhibit a broad single peak. Depending on the mean particle diameter the peak width varies. For smaller particles, the energetic width of the peak is broader. This observation strongly suggests that the peak broadening is caused by the particle size distribution [6].

Present work deals with the particle size distribution of semiconductor quantum dot effect on the spectral characteristics of luminescence and model the theory using MATLAB and MATHEMATICA 5.2 as a tool for various materials.

THEORETICALLY

If the particle size is not uniform but has a distribution the observed spectrum of the quantum dot system can be taken as the superposition of the contribution from each individual particle. As each particle would exhibit a peak at a position dictated by its diameter, this would result in an inhomogeneous broadening of PL spectrum. The overall line shape can be obtained by integrating $g_{qd}(E,R)$ over size distribution $P(R)$ as[27]

$$G(E) = \int P(R) \cdot g_{qd}(E, R) dR \quad 1.1$$

where $P(R)$ is probability of particle of radius R .

For most common distribution, $P(R)$ is taken to be Gaussian

$$\text{given by } P(R) = \frac{1}{\sigma_R \sqrt{2\pi}} \exp \left[-\frac{(R - R_0)^2}{2\sigma_R^2} \right] \quad 1.2$$

where R_0 is mean radius, σ_R standard deviation [27] given by

$$\sigma_R = \frac{\sum(R-R_0)^2}{n-1}$$

with relative standard deviation (dispersion %) = $\frac{\sigma_R \times 100}{R_0}$
 and $g_{qd}(E, R)$ is the line shape function for nano-particles
 of radius R in the range

$R = R_0 \pm 3\sigma_R$ [27] given by

$$g_{qd}(E, R) = \frac{A}{\Gamma\sqrt{2\pi}} \exp\left[-\frac{(E - E(R))^2}{2\Gamma^2}\right] \quad 1.3$$

$E(R)$ is the effective band gap energy between valence band edge and conduction band edge of Q-dots and can be calculated using equation

$$E(R) = E_0 + \frac{\hbar^2 \pi^2}{2} \left(\frac{1}{m_e^*} + \frac{1}{m_h^*} \right) \frac{1}{R^2} - \frac{1.8e^2}{\epsilon_2} \frac{1}{R} \quad 1.4$$

the above equation for ZnSe and CdSe Q dots with mean radius R_0 is given by

$$E(R_0) = 2.81 + \frac{2.43}{(R_0)^2} - \frac{0.454}{R_0} \quad \text{(ZnSe)} \quad 1.5$$

$$E(R_0) = 1.71 + \frac{3.73}{(R_0)^2} - \frac{0.278}{R_0} \quad \text{(CdSe)} \quad 1.6$$

Line shape function for Poly-disperse ZnSe

1. For 1nm monodisperse ZnSe ;dispersion factor $\sigma_R/R_0 =$ zero. Hence the expression for line shape function of mean radius R_0 is computed by equation:

$$g_{qd}(E, R_0) = \frac{A}{\Gamma\sqrt{2\pi}} \exp\left[-\frac{(E - E(R_0))^2}{2\Gamma^2}\right] \quad 1.7$$

2. For 1nm Polydisperse ZnSe; $R_0=1\text{nm}$

a. Dispersion factor $\sigma_R/R_0 = 5\%$

Standard deviation $\sigma_R = .05$ and $R = .85$ to 1.15

b. For Dispersion factor $\sigma_R/R_0 = 10\%$

Standard deviation $\sigma_R = .1$ and $R = .7$ to 1.3

c. For Dispersion factor $\sigma_R/R_0 = 15\%$

Standard deviation $\sigma_R = .15$ and $R = .55$ to 1.45

d. For Dispersion factor $\sigma_R/R_0 = 20\%$

Standard deviation $\sigma_R = .2$ and $R = .4$ to 1.6

e. For Dispersion factor $\sigma_R/R_0 = 25\%$

Standard deviation $\sigma_R = .25$ and $R = .25$ to 1.75

Now $E(R)$ for polydisperse ZnSe is expressed using equation 1.4. as

$$E(R) = 2.81 + \frac{2.43}{(R)^2} - \frac{0.454}{R} \quad 1.8$$

Hence using the above parameters line shape function $G(E)$ for polydisperse ZnSe with $R_0= 1\text{nm}$ for different dispersions can be calculated and plotted for different values of E using equation . 1.1

The Modeling for spectral characteristics of polydisperse ZnSe for $R_0=1\text{nm}$ and dispersion 0,5,10,15 and 25%

(* ***** *)

(* 1. 0% Dispersion ZnSe $r_0=1\text{nm}$ 0; *)

$r_0=1;$

(* e=2 *)

$\tau=.1;$

$a=.25;$

$\epsilon_r=4.77;$

$g_{qd0}=(a/(\tau*\text{Sqrt}[2*Pi]))*\text{Exp}[-.5*((e-\epsilon_r)/\tau)^2]$

$\text{plot1nm0} = \text{Plot} [g_{qd0} , \{e,0,7\} , \text{PlotRange} \rightarrow \{0,1.0\} , \text{PlotLabel} \rightarrow \text{"Graph of 1nm ZnSe for 0% Dispersion"} , \text{PlotStyle} \rightarrow \{\text{RGBColor}[1,0,1]\}] ;$

(* ***** *)

(* 1. 5% Dispersion ZnSe $r_0=1\text{nm}$; *)

$r_0=1;$

(* e=2 *)

$\tau=.1;$

$a=.25;$

$\text{sigmar1}=.05;$

$\epsilon_r=2.81+2.42/r^2-.454/r;$

$\text{pr1}=(1/(\text{sigmar1}*\text{Sqrt}[2*Pi]))*\text{Exp}[-.5*((r-r_0)/\text{sigmar1})^2]$

$g_{qd1}=(a/(\tau*\text{Sqrt}[2*Pi]))*\text{Exp}[-.5*((e-\epsilon_r)/\tau)^2]$

$\text{Table}[\text{NIntegrate}[\text{pr1}*g_{qd1},\{r,.85,1.15\}],\{e,3.5,5.5,.1\}]$

$\text{plot1nm5} = \text{Plot} [\text{NIntegrate}[\text{pr1}*g_{qd1},\{r,.85,1.15\}] , \{e,0,7\} , \text{PlotRange} \rightarrow \{0,1.0\} , \text{PlotLabel} \rightarrow \text{"Graph of 1nm ZnSe for 5% Dispersion"} , \text{PlotStyle} \rightarrow \{\text{RGBColor}[0,1,1]\}] ;$

(* ***** *)

(* 1. 10% Dispersion ZnSe $r_0=1\text{nm}$; *)

$r_0=1;$

(* e=2 *)

```
tau=.1;
a=.25;
sigmar2=.1;
er=2.81+2.42/r^2-.454/r;
pr1=(1/(sigmar2*Sqrt[2*Pi]))*Exp[-.5*((r-r0)/sigmar2)^2]
gqd1=(a/(tau*Sqrt[2*Pi]))*Exp[-.5*((e-er)/tau)^2]
Table[NIntegrate[pr1*gqd1,{r,.7,1.3}],{e,3.5,5.5,.1}]
plot1nm10 = Plot [ NIntegrate[pr1*gqd1,{r,.7,1.3}], {e,0,7} ,
PlotRange-> {0,1.0} , PlotLabel -> "Graph of 1nm ZnSe for
10% Dispersion" , PlotStyle->{RGBColor[0,1,0]} ] ;
```

```
(* ***** *)
(* 1. 15 % Dispersion ZnSe r0=1nm ; *)
r0=1;
(* e=2 *)
tau=.1;
a=.25;
sigmar3=.15;
er=2.81+2.42/r^2-.454/r;
pr1=(1/(sigmar3*Sqrt[2*Pi]))*Exp[-.5*((r-r0)/sigmar3)^2]
gqd1=(a/(tau*Sqrt[2*Pi]))*Exp[-.5*((e-er)/tau)^2]
Table[NIntegrate[pr1*gqd1,{r,.55,1.45}],{e,2.5,6.5,.1}]
plot1nm15 = Plot [ NIntegrate[pr1*gqd1,{r,.55,1.45}],
{e,0,7} , PlotRange-> {0,1.0} , PlotLabel -> "Graph of 1nm
ZnSe for 15% Dispersion" , PlotStyle->{RGBColor[0,0,1]} ] ;
```

```
(* ***** *)
(* 1. 20 % Dispersion ZnSe r0=1nm ; *)
r0=1;
(*e=2*)
tau=.1;
a=.25;
sigmar4=.2;
er=2.81+2.42/r^2-.454/r;
pr2=(1/(sigmar4*Sqrt[2*Pi]))*Exp[-.5*((r-r0)/sigmar4)^2]
gqd2=(a/(tau*Sqrt[2*Pi]))*Exp[-.5*((e-er)/tau)^2]
Table[NIntegrate[pr2*gqd2,{r,.4,1.6}],{e,2.5,6,.1}]
plot1nm20 = Plot [ NIntegrate[pr2*gqd2,{r,.4,1.6}], {e,0,7} ,
PlotRange-> {0,1.0} , PlotLabel -> "Graph of 1nm ZnSe for
20% Dispersion" , PlotStyle->{RGBColor[1,0,0]} ]
```

```
(* ***** *)
(* 1. 25 % Dispersion ZnSe r0=1nm ; *)
r0=1;
(*e=2*)
tau=.1;
```

```
a=.25;
sigmar5=.25;
er=2.81+2.42/r^2-.454/r;
pr=(1/(sigmar5*Sqrt[2*Pi]))*Exp[-.5*((r-r0)/sigmar5)^2]
gqd=(a/(tau*Sqrt[2*Pi]))*Exp[-.5*((e-er)/tau)^2]
Table[NIntegrate[pr*gqd,{r,.25,1.75}],{e,2,7,.1}]
plot1nm25 = Plot [ NIntegrate[pr*gqd,{r,.25,1.75}], {e,0,7} ,
PlotRange-> {0,1.0} , PlotLabel -> "Graph of 1nm ZnSe for
25% Dispersion" , PlotStyle->{RGBColor[1,1,2]} ]
```

```
(* ***** *)
Show [ plot1nm0 , plot1nm5 , plot1nm10 , plot1nm15 ,
plot1nm20 , plot1nm25 , DefaultFont->{"Helvetica",14} ,
PlotLabel -> "Graph of 1nm ZnSe for 0%,5%,10%,15%,20%
and 25% Dispersion" , Frame-> True , FrameLabel-
->{"Energy [eV]" , "Intensity [arb.units]" } ] .
```

Using similar calculations performed for 1nm polydisperse ZnSe we can model the program for spectral characteristics of polydisperse ZnSe for $R_0 = 1.5, 2, 3$ nm by substituting different values of σ_R and R for varying R_0 keeping all other parameters same. Further applying the same theoretical calculations we can also model the program for CdSe Q.dots.

The spectral characteristics for 1, 1.5, 2, 3nm ZnSe polydisperse quantum dots is shown below:

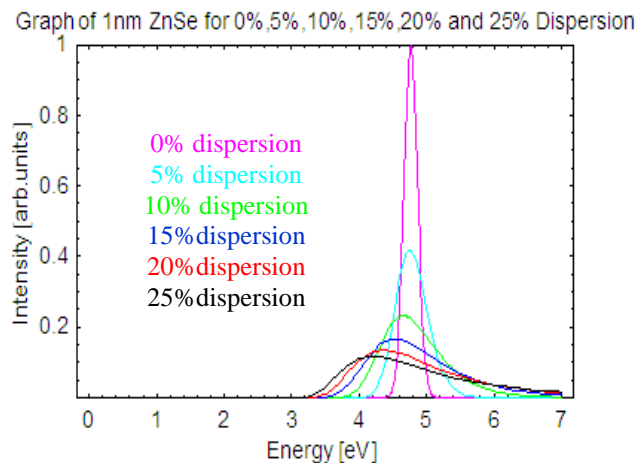


Figure 1: 1nm polydisperse ZnSe Q.dots.

Graph of 1.5nm ZnSe for 0%,5%,10%,15%,20% and 25% Dispersion

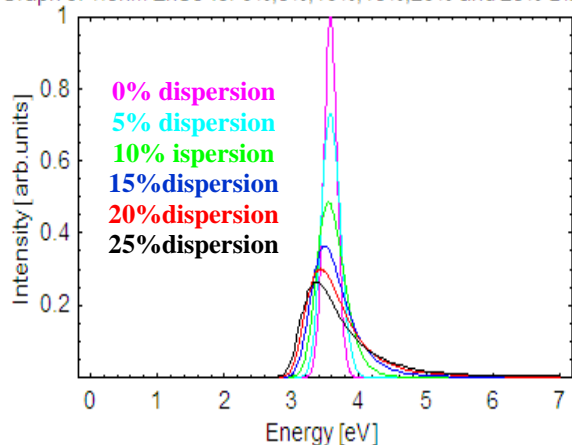


Figure 2: 1.5nm polydisperse ZnSe Q. dots.

Graph of 2nm ZnSe for 0%,5%,10%,15%,20% and 25% Dispersion

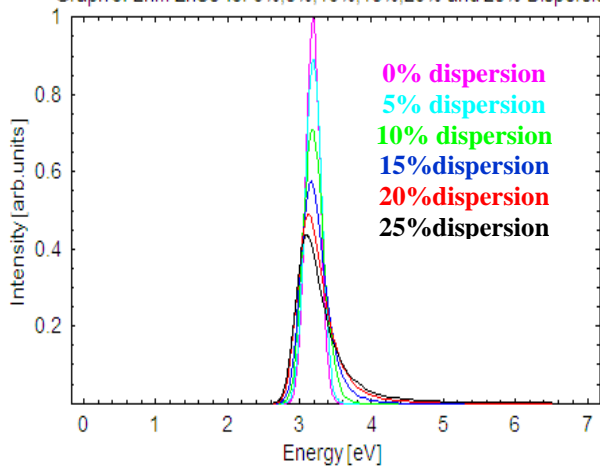


Figure 3: 2nm polydisperse ZnSe quantum dots.

Graph of 3nm ZnSe for 0%,5%,10%,15%,20% and 25% Dispersion

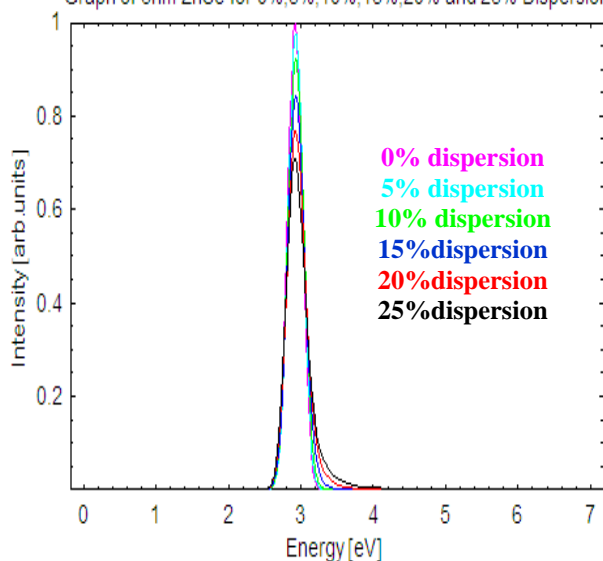


Figure 4: 3nm polydisperse ZnSe quantum dots.

Similarly we can also get the spectral characteristics of CdSe Q. dots for 1, 1.5,2, 3nm.

Table 1: Calculated values of PL, peak photon energy, intensity & width of poly disperse- ZnSe quantum dots

Average Size (nm)	Dispersion (%)	Peak at (eV)	Intensity (arb. Units)	FWHM (eV)
1	0	4.78	1	0.12
	5	4.7	0.408	0.57
	10	4.6	0.23	1.1
	15	4.5	0.165	1.4
	20	4.4	0.133	1.65
	25	4.2	0.116	1.8
1.5	0	3.58	1	0.12
	5	3.5	0.724	0.45
	10	3.5	0.47	0.48
	15	3.4	0.364	0.52
	20	3.3	0.298	0.55
	25	3.2	0.262	0.6
2	0	3.19	1	0.12
	5	3.1	0.88	0.2
	10	3.1	0.704	0.26
	15	3.1	0.558	0.32
	20	3	0.482	0.35
	25	2.9	0.436	0.4
3	0	2.93	1	0.12
	5	2.9	0.938	0.15
	10	2.9	0.882	0.17
	15	2.9	0.814	0.2
	20	2.9	0.752	0.21
	25	2.9	0.701	0.23

The overall comparable observations for both quantum dots are as follows:

- 1) The maximum intensity observed for mono disperse ZnSe of radii 1nm ,1.5nm, 2nm and 3nm is at 4.78, 3.58, 3.19 and 2.93eV, where as for mono disperse CdSe of radii 1nm,1.5nm, 2nm and 3nm is at 5.18, 3.20, 2.50 and 2.03eV, which clearly predicts blue shift with the decrease in size in both cases. But as compared to ZnSe we observe that except for monodisperse particle, the intensity for CdSe Q dots is least for all sizes with dispersion.

- 2) As the percentage of dispersion increases the peak intensity decreases thereby increasing the FWHM for a fixed average size. But the FWHM here is same as in case of for all dispersion and sizes as the size distribution factor is independent of intrinsic line width.
- 3) Red shift in the PL peaks is obtained when the size distribution increases from 0 to 25% for all average sizes.
- 4) The intensity of PL peaks decreases with the increase in size distribution for 1nm, 1.5nm, 2nm and 3nm but the magnitude of line shape intensity increases to maximum when the size advances towards higher radii.
- 5) Inhomogeneous broadening is increased with increase in the size distribution but the broadening is reduced when the average size increases from 1nm to 3nm.

CONCLUSION

The effect of particle size distribution on PL spectra of different QD sizes of material is investigated using modeling. Ravindran et al [27] have attempted to describe broadening of emission peaks of CdS nanoparticles due to size distribution. They have also shown asymmetric broadening and slight red shift of PL peak due to size distribution. Experimentally they have obtained two PL peaks one from band edge luminescence and the other due to recombination at defects. Analysis by curve fitting is used to estimate average particle size and standard deviation of size distribution. The particle size is in good agreement with that obtained from XRD (~4nm) and the width of PL peak shows 6% polydispersion. This agrees our theoretical analysis.

Kurrey et al.[28] studied that nanophosphors synthesized through solgel method with narrow size distribution, showed broadening in the PL peaks. The size and morphology of the prepared ZnO nanophosphors has been confirmed through XRD and SEM. The prepared ZnO nanophosphor has enough potential for optoelectronic applications.

Alipieva et al [29] have reported influence of quantum dots size dispersion on the fluorescence spectrum of CdSe/ZnS quantum dots. They have prepared the QDs of 2.5nm and 5nm in porous matrix in various ratios and investigated their fluorescence by exciting with 405nm semiconductor laser. It was found that 2.5nm QDs emit light in green region at ~540nm and 5nm QDs emit at ~620nm. When QDs of both sizes were mixed in different ratios 1:1, 1:2, and 1:3, it was observed that luminescent of smaller QDs has quenched by larger QDs, though line shape function is nearly Gaussian only. This indicates that if there is size distribution in nanoparticles samples the light emission will be more from the larger QDs, shifting the net peaks towards higher wave length or lower energies. Our theory also predicts the same as seen from figures 1 to 4.

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