

# CHAPTER IV

## RESULTS & DISCUSSION



Optical characterization of linearly ordered QD structure in the presence of an applied electric field is thoroughly mentioned in this chapter. Calculations of the two-dimensional aligned QDs (by using the Schrödinger equation) with the two-dimensional electric field (by using the Laplace's equation) and manipulate them for obtaining the solution of the system. The investigation is demonstrated by varying some associated parameters, for instance, QD size, interdot spacing, number of QDs, electric field's direction, and amplitude of applied field in order to observe their optical properties and compare the results with the previous cases (elongated QD and aligned QDs in absence of electric field). These steps allow the calculation results to show how the QCSE affects the polarization anisotropy, which is related to how the electroabsorption phenomenon acts upon nanostructures.

In essence, the above mechanisms are expected to be one of the valuable information of well-established theories aiming to develop powerful nanophotonic devices in the short run. Finally, an overall discussion is addressed to summarize the results, which are then considered for organizing an intellectual thought process leading to the absolute conclusion in the thesis work.

### 4.1 QUANTUM DOTS IN TWO-DIMENSIONAL ELECTRIC FIELD SYSTEM

To begin with, recalling from the time-independent Schrödinger equation (Eq. (3.1)) in a two-dimensional system, now we take an external electric field into account (Eq. (3.23)) of QD system. With these equations, to obtain the Hamiltonian operator  $H(E)$  in the presence of the electric field  $E$ , it is necessary to add the Stark energy (under the applied field distributing in 2-D) terms,  $qE(x, y)$ , into potential energy side of the Hamiltonian. The effective Hamiltonian of the electron and hole can be written as follows:

For the electron,

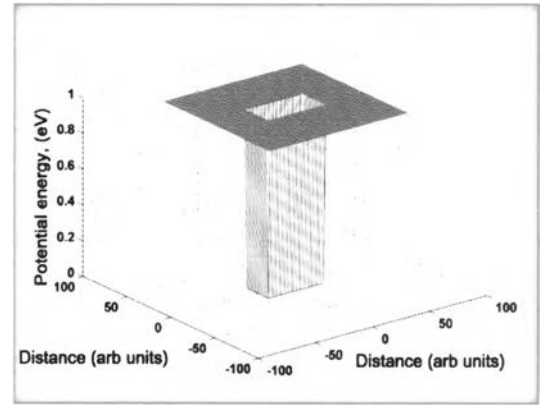
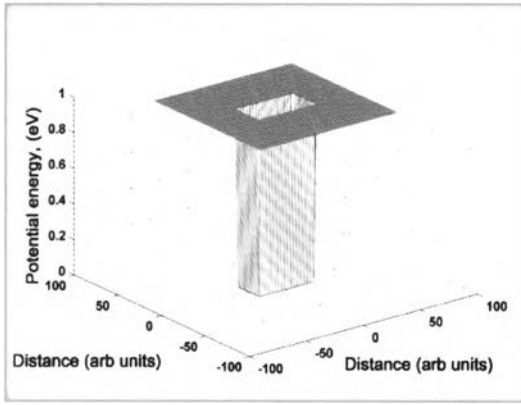
$$H_e \psi_e(x, y) = \left[ -\frac{\hbar^2}{2m_e^*} \frac{\partial^2}{\partial x^2} + V_e(x, y) - qE_x(x, y) - qE_y(x, y) \right] \psi_e(x, y) = E \psi_e(x, y) \quad (4.1)$$

and for the hole,

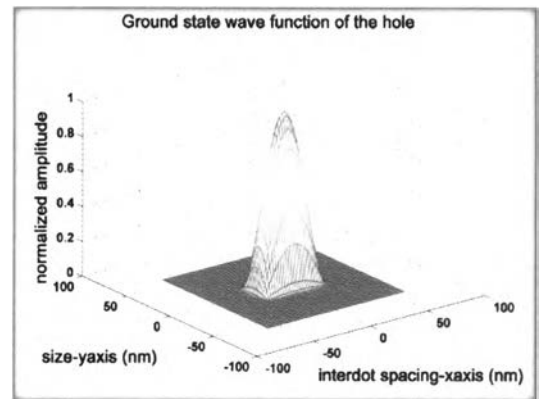
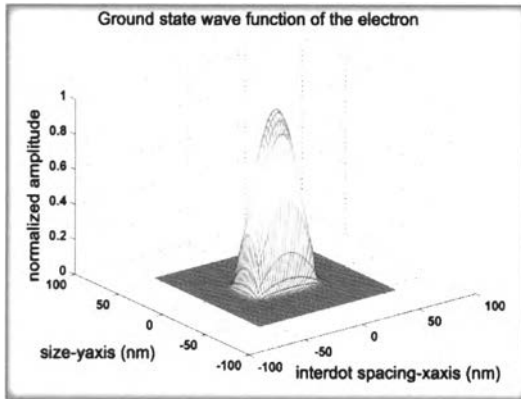
$$H_h \psi_h(x, y) = \left[ -\frac{\hbar^2}{2m_h^*} \frac{\partial^2}{\partial x^2} + V_h(x, y) + qE_x(x, y) + qE_y(x, y) \right] \psi_h(x, y) = E \psi_h(x, y) \quad (4.2)$$

where  $q$  is the electronic charge. The subscripts  $e$  and  $h$  represent the electron and the hole, respectively.  $m_e^*$  and  $m_h^*$  are effective masses of the electron and the hole, respectively.  $V_e(x, y)$  and  $V_h(x, y)$  are the band offset, or the difference between the bandgap energy of the InAs quantum dots and the GaAs surrounding matrix, which is given in the ratio of 0.7 : 0.3, respectively (as mentioned in chapter 3).  $(x, y)$  is the Cartesian coordinates corresponding to the electric field at that position.  $E_x$  and  $E_y$  are electric field along the  $x$  (parallel to the alignment of ordered QDs or horizontal axis) and  $y$  (normal to the alignment of ordered QDs or perpendicular axis) direction, respectively. Note that the negative sign in front of the electronic charge  $q$  in case of the electron Hamiltonian comes from the scalar value which is necessary to be significantly considered.

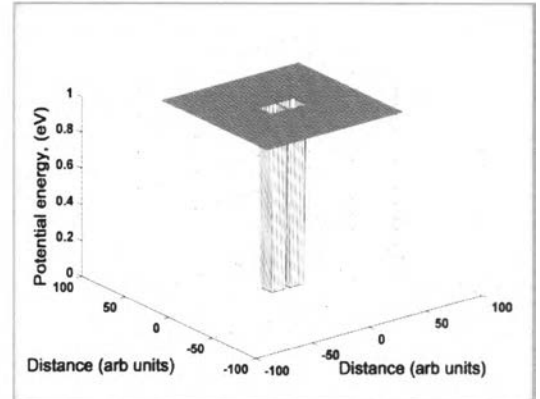
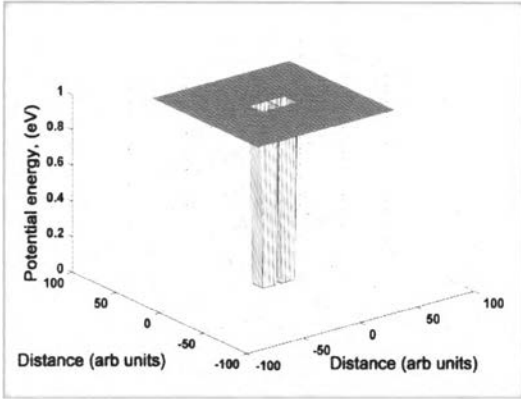
Solving both Eq. (4.1) and Eq. (4.2) numerically, we obtain the electron and hole wavefunctions of the system, respectively. The stability of a finite difference scheme can work depending on the value of each mesh ( $h$ ), which is consistent with the local truncation. On the other hand, the limit of the local truncation error is zero as  $h$  approaches zero (here, we used the number of mesh points of 9801x9801 as described in chapter 3 to give the highest possible accuracy). Hence, we use these to calculate the optical intensity (Eq. (3.7)), as a result, the degree of polarization in case of single (Eq. (3.8)) and aligned QDs (Eq. (3.13)) can be obtained.



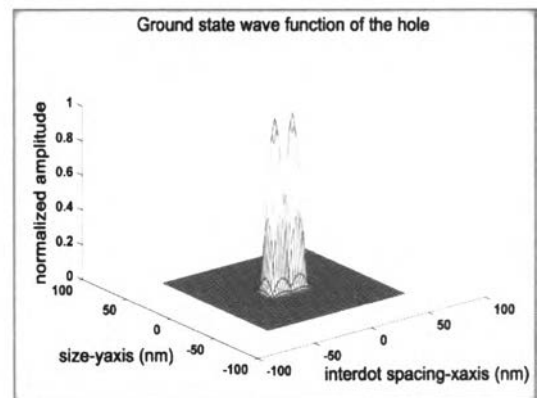
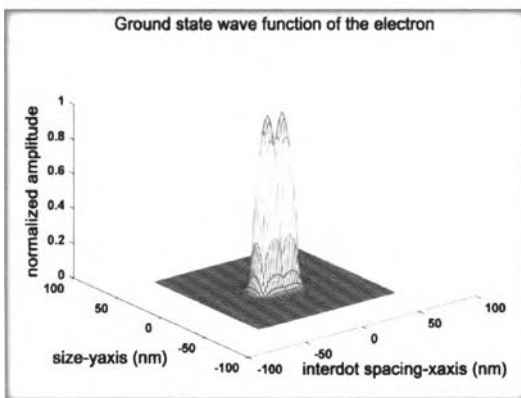
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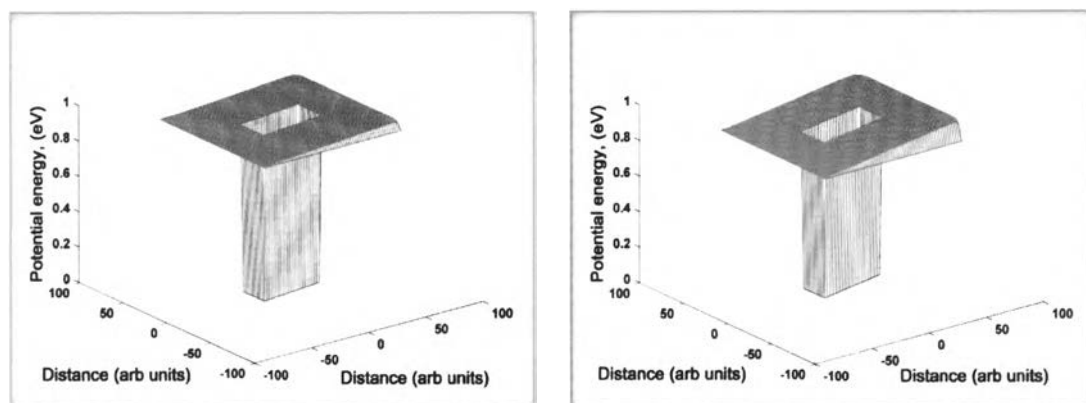
(d)

**Figure 4.1** (a) The normalized conduction band (CB) structure (left) and valence band (VB) structure (right) of elongated QD whose size is  $24 \times 48 \text{ nm}^2$ . (b) The ground-state electron (left) and hole (right) wavefunctions related to CB and VB, respectively. (c) The normalized conduction band (CB) structure (left) and valence band (VB) structure (right) of binary QDs, each of size  $12 \times 12 \text{ nm}^2$  with interdot spacing of 2 nm. (d) The ground-state electron (left) and hole (right) wavefunctions related to CB and VB, respectively. These results are under the condition of no electric field in any directions ( $E_x$  and  $E_y = 0 \text{ V/nm}$ ).

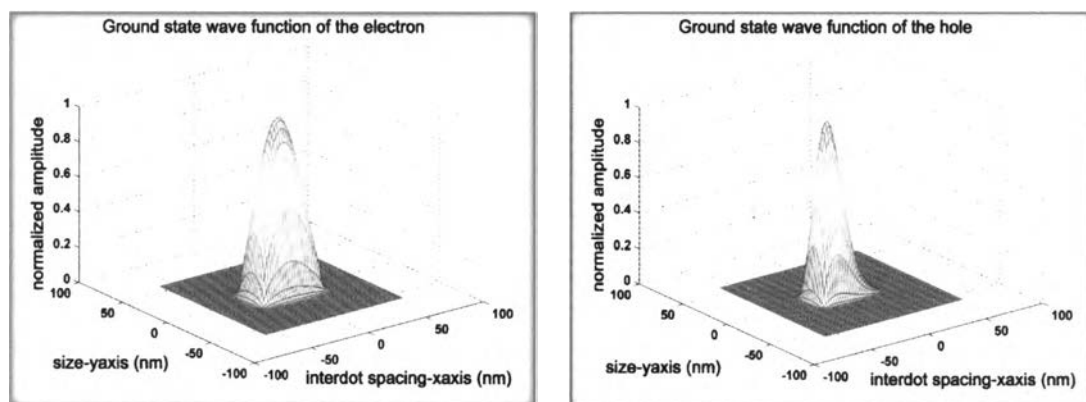
#### 4.1.1 Influence of the Electric Field Direction on the Polarization Degree

As a preliminary calculation, we emphasize on the study of the influence of the electric field direction on the polarization degree. A single QD, whose size in the x and the y direction fixed to 48 and 24 nm, respectively, was prepared for calculating the PD values. Besides, the two and four aligned QDs (in the x direction) were also put into practice by using  $12 \times 12 \text{ nm}^2$  isotropic QDs with interdot spacing maintained at 2 nm. **Figure 4.1** shows the band structure, the ground-state electron, and the ground-state hole wavefunctions related to the QDs. At the initial state, there is no external energy performing on the system (in absence of an applied voltage), an equilibrium situation is then revealed by the normality of potential well for both conduction band and the valence band (**Figure 4.1 (a) and (c)**) and the symmetric distribution of the electron and hole wavefunctions (**Figure 4.1 (b) and (d)**). The width of electron distribution function is also larger than that for the hole.

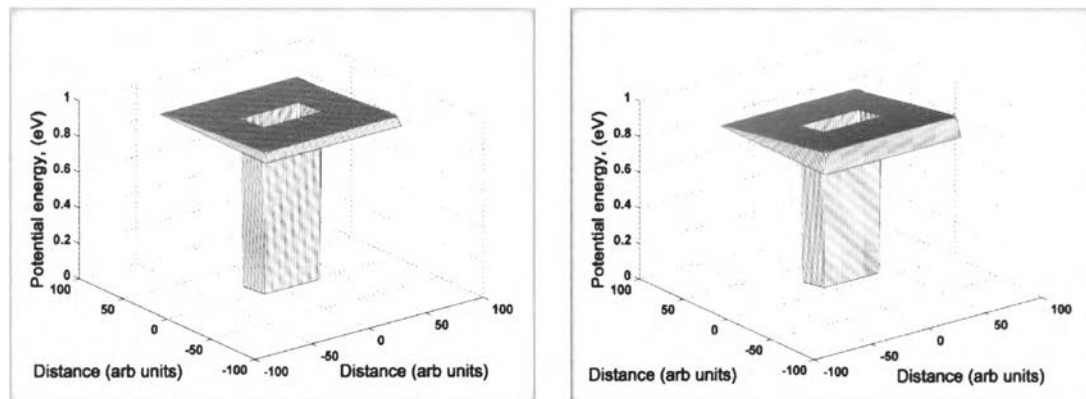
When the QD group is subject to the external applied voltage, the system is no longer in equilibrium. In case the bias voltage of 0.08 V was applied along the x direction (direction of QD elongation/alignment) producing the electric field parallel



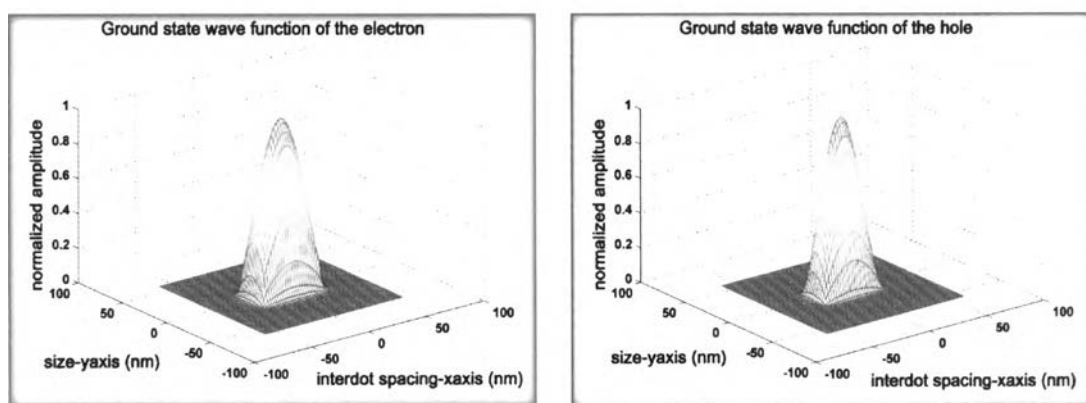
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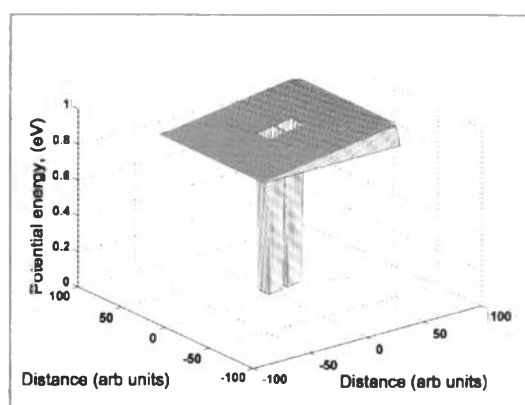
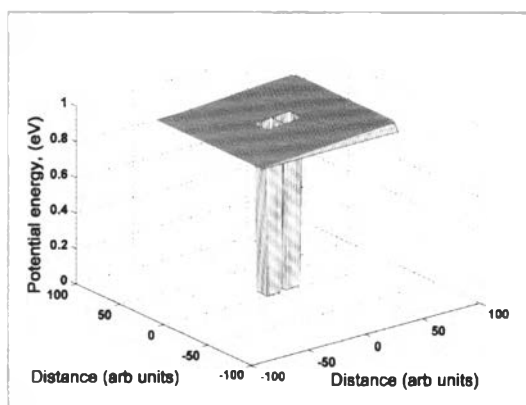


(d)

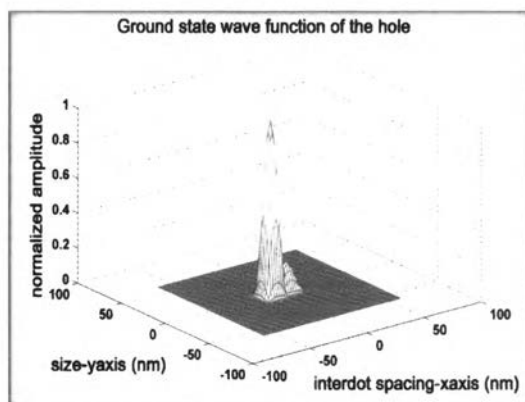
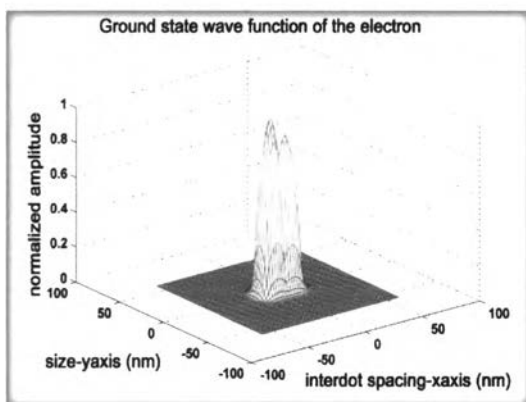
**Figure 4.2** (a) The normalized conduction band (CB) structure (left) and valence band (VB) structure (right) of elongated QD whose size is  $24 \times 48 \text{ nm}^2$ . The results are for the applied voltage with a magnitude of  $0.008 \text{ V}$  parallel to the  $x$  direction or direction of QD elongation (producing an electric field in the same direction ( $E_x > 0, E_y = 0 \text{ V/nm}$ )). (b) The ground-state electron (left) and hole (right) wavefunctions for the CB and VB, respectively. (c) The normalized conduction band (CB) structure (left) and valence band (VB) structure (right) when the elongated QD is under the applied voltage with a magnitude of  $0.008 \text{ V}$  parallel to the  $y$  direction or perpendicular direction ( $E_y > 0, E_x = 0 \text{ V/nm}$ ). (d) The ground-state electron (left) and hole (right) wavefunctions for the CB and VB, respectively.

to that direction (Note that these parameters are represented as  $V_x$  and  $E_x$ , respectively, and will be used hereafter), several phenomena happen, as shown in **Figure 4.2** and **Figure 4.3**. When the applied field acts on the system, the conduction band and the valence band structure become tilted down along the electric field direction (**Figure 4.2 (a)** and **Figure 4.3 (a)**) [126]. It separates the electron and the hole (which were originally localized) and moves them apart from each other. In other words, the electron and hole wavefunctions are separated and pushed toward opposite sides of the well, that is, the positions of the distribution maxima of the electron and hole shift away from the center: the electron (hole) shifts toward the negative (positive)  $x$  direction or the direction of electric field distribution (see **Figure 3.9**). So excitons in QDs are “polarized” by the applied electric field.

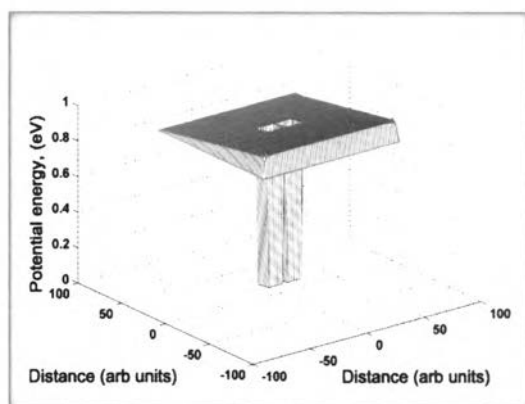
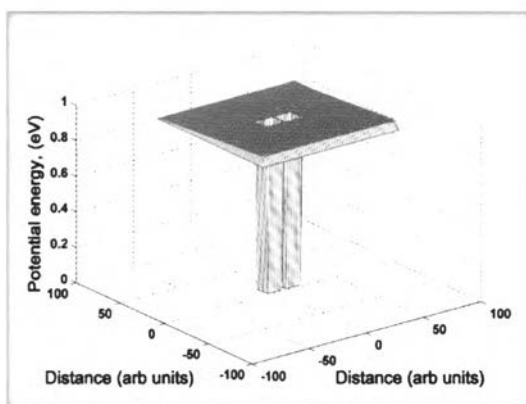
Furthermore, the width of the distribution function for the electron under the electric field is larger than that of the hole due to the smaller effective mass (larger kinetic energy) of the electron than that of the hole, which causes the electron to distribute farther away from the center than the hole, but the hole wavefunction affected by the electric field is more distorted than the electron wavefunction (**Figure 4.2 (b)**), which is obviously seen when the number of QDs



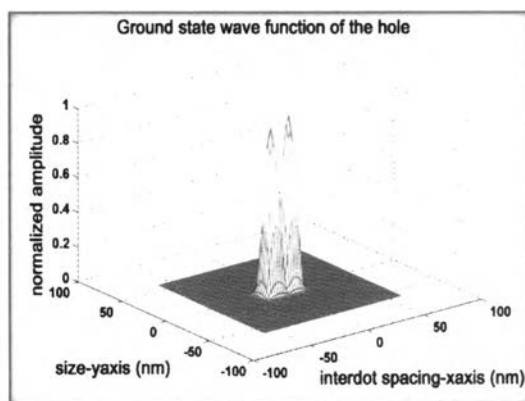
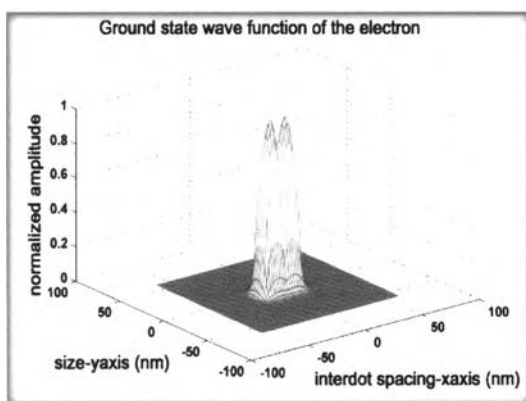
(a)



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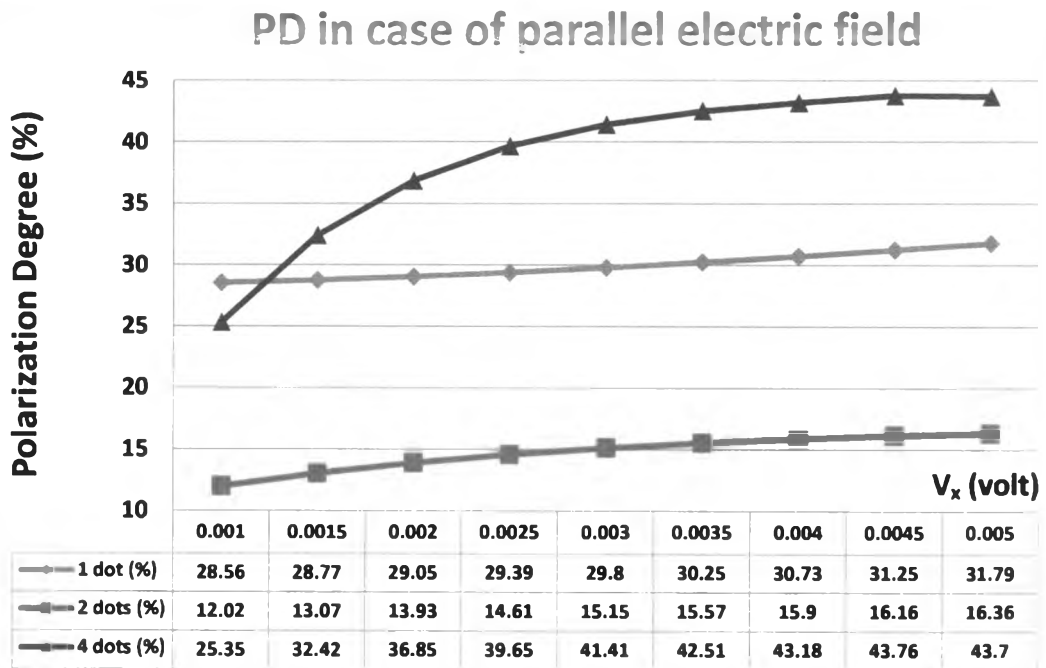
**Figure 4.3** (a) The normalized conduction band (CB) structure (left) and valence band (VB) structure (right) of binary QDs, each of size  $12 \times 12 \text{ nm}^2$  with interdot spacing of 2 nm. The results are under the applied voltage with a magnitude of 0.008 V parallel to the x direction or aligned QD direction (producing an electric field in the same direction ( $E_x > 0, E_y = 0 \text{ V/nm}$ )). (b) The ground-state electron (left) and hole (right) wavefunctions related to CB and VB, respectively. (c) The normalized conduction band (CB) structure (left) and valence band (VB) structure (right) when the binary QDs is the under the applied voltage with a magnitude of 0.008 V parallel to the y direction ( $E_y > 0, E_x = 0 \text{ V/nm}$ ). (d) The ground-state electron (left) and hole (right) wavefunctions for CB and VB, respectively.

increases (**Figure 4.3 (b)**). Therefore, it is easier to observe the change in hole wavefunction than the electron's, and it is necessary to change the shape of the hole wavefunction in order to control the optical polarization property [197].

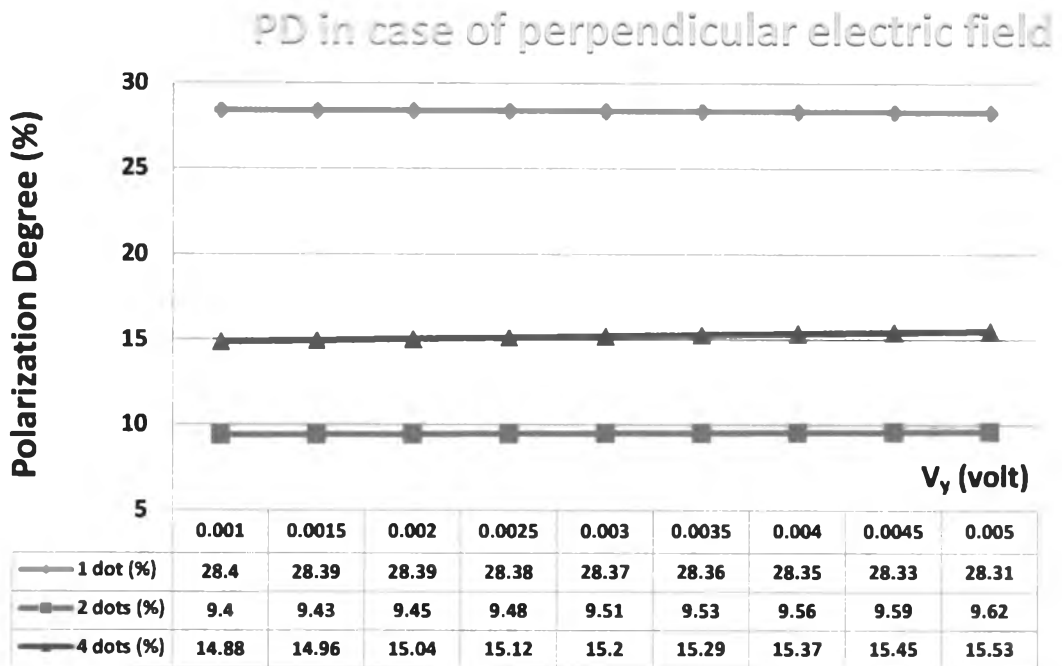
For the case of bias voltage applied along the y direction or  $V_y$  (perpendicular to the direction of QD elongation/alignment), it is seen that both electron and hole wavefunctions maintain their spatial symmetry, as shown in **Figure 4.2 (d)** and **Figure 4.3 (d)** even though the band tilting occurs for the conduction and valence bands as in the previous case.

Consider the polarization degree of these three QD cases plotted in **Figure 4.4** under the applied voltage. It was found that the polarization degree of elongated QD, binary QDs, and four QDs increased with increasing applied voltage  $V_x$  (**Figure 4.4 (a)**) whereas they were almost unchanged when the voltage was applied along the y direction. All these results confirm that the perpendicular electric field weakly affects the distribution function of excitons. As a result, the electric field  $E_y$  does not significantly change the degree of polarization since the applied field is unable to





(a)

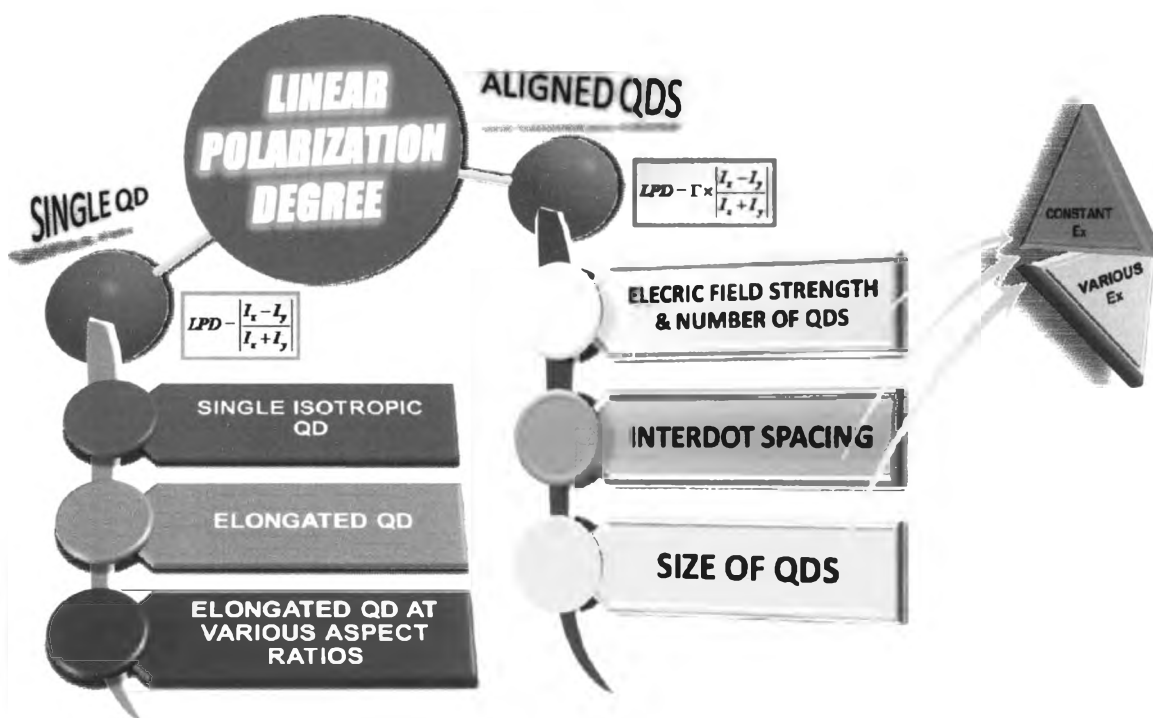


(b)

**Figure 4.4** The polarization degree of elongated QD ( $24 \times 48 \text{ nm}^2$ ), binary QDs, and four QDs ( $12 \times 12 \text{ nm}^2$ ) under the applied voltage in the (a) x direction (parallel to the alignment of QD) and (b) y direction (normal to the alignment of QD) with the range between 0.001 and 0.005 V.

cause any difference between the transition probability both two directions. In contrast, their PD values are strongly affected by the parallel electric field which can be described as follows. The elongated or the aligned QD structure not only enables carriers to move along these directions mentioned above, but also increases the transition probability in those directions, leading to the increase of LPD.

The existence of applied electric field along the direction of QD alignment or extending direction of single QD will support the carrier transition to move more comfortably along these directions by tunneling effect (which will be explained in next section) on account of band tilting which allows for easier absorption process, then producing more and more coherently light emission. All the results refer to QCSE and are consistent with the calculations previously reported in [134, 146, 152, 197]. According to the reasons clearly seen from the influence of electric field's direction, we only focus on the electric field in the direction of QDs alignment or applied voltage  $V_x$  from now on.



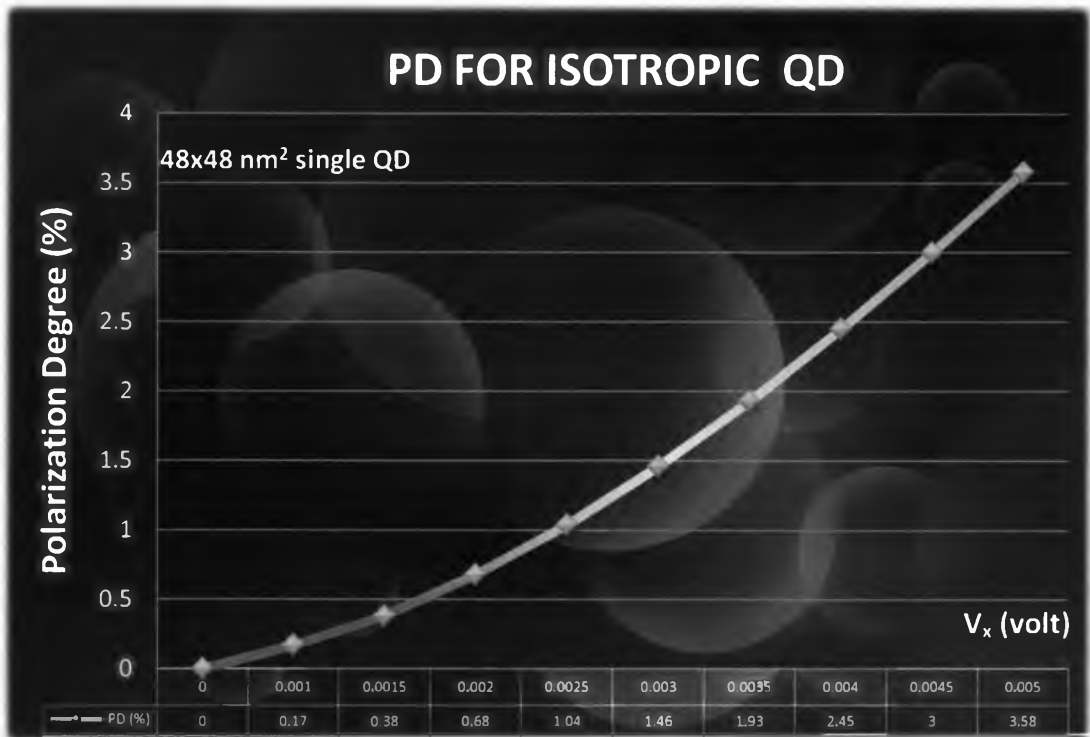
**Figure 4.5** Diagram of investigation of linear polarization degree on single and aligned QD structure in presence of an applied electric field.

## 4.2 LPD CHARACTERISTIC OF SINGLE QD UNDER THE ELECTRIC FIELD

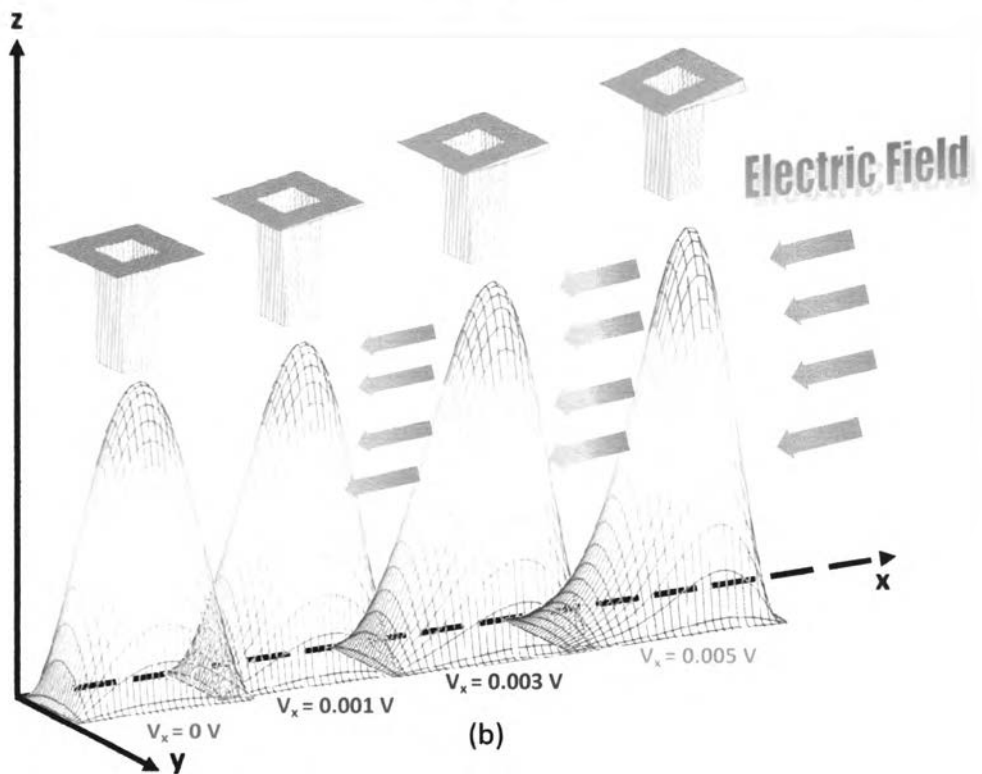
In this section, we continue studying the polarization degree of single QD in the presence of an electric field. We investigated by varying QD sizes and applied field. This enables us to evaluate the optical polarization degree from the electron and hole wavefunctions in case of both the asymmetric structure (elongated QD) and the influence of electric field strength, and compare the results with the former cases (**section 3.1.4.2.1**) so that consideration of how the calculated results agree or contradict with each other. The detailed physics of the quantum confined Stark shift of excitons was simultaneously given to support the calculated data and obtain the exact information of single QDs. Moreover, this information will be collected as a database for comparison with the case of aligned QD system.

### 4.2.1 Increase of Polarization Degree of Isotropic QD with Applied Electric Field

Consider the polarization degree of single QD with an isotropic shape ( $48 \times 48 \text{ nm}^2$ ), as shown in **Figure 4.6 (a)**. It manifests a non-polarized value in a zero electric field due to spatially symmetric shape in both x and y directions causing an unchanged PD of the system (as mentioned in **section 3.1.4.2.1**). When an electric field is applied (created by  $V_x$ ), PD simply increases as the electric field is increased. **Figure 4.6 (b)** displays the hole wavefunction concentrated along the x direction as  $V_x$  is non-zero (electric field is now applied at the  $-x$  direction). The distribution maxima move toward the  $+x$  ( $-x$ ) direction for hole (electron) as the electric field increases. Both the electron and hole tend to distribute themselves near the surface under the strong electric field. The distribution function of the hole in this case is still narrower than that of the electron (as discussed previously in **section 4.1.1**). The hole wavefunction finally moves to the maximum position when  $V_x$  reaches to 0.005 V (as previously described, we display only the hole wavefunction because it can be



(a)



**Figure 4.6** (a) The polarization degree of single QD with an isotropic shape (48x48 nm<sup>2</sup>) vs. applied voltage  $V_x$  varied from 0 to 0.005 V. (b) The ground-state hole wavefunction under the  $V_x$  of 0, 0.001, 0.003, and 0.005 V, respectively.

more easily observed for the change of its shape than that of the electron). This effect is attributed to the influence of the electric field which acts on the QD by pushing the electron and the hole to the opposite directions along the x axis and the spatial expansion of electron and hole wavefunctions (initially completely symmetric) are more distributed along the x direction (but spatially localized), leading to higher transition probability in the x direction than the y direction. So  $I_x$  becomes larger than  $I_y$ , resulting in a finite value for the degree of polarization. Increasing the applied field causes the potential well to tilt down, which allows the valence electron to tunnel more easily through a triangular barrier of height  $E_g$  to the conduction band. In addition, the appropriate strength of electric field can assist the incoming photon whose energy is less than the bandgap energy ( $\hbar\omega < E_g$ ) to be absorbed. In other words, the probability of carriers tunneling out of the well also increases, which in turn decreases in carrier lifetimes, and then the degree of optical polarization increase as well.

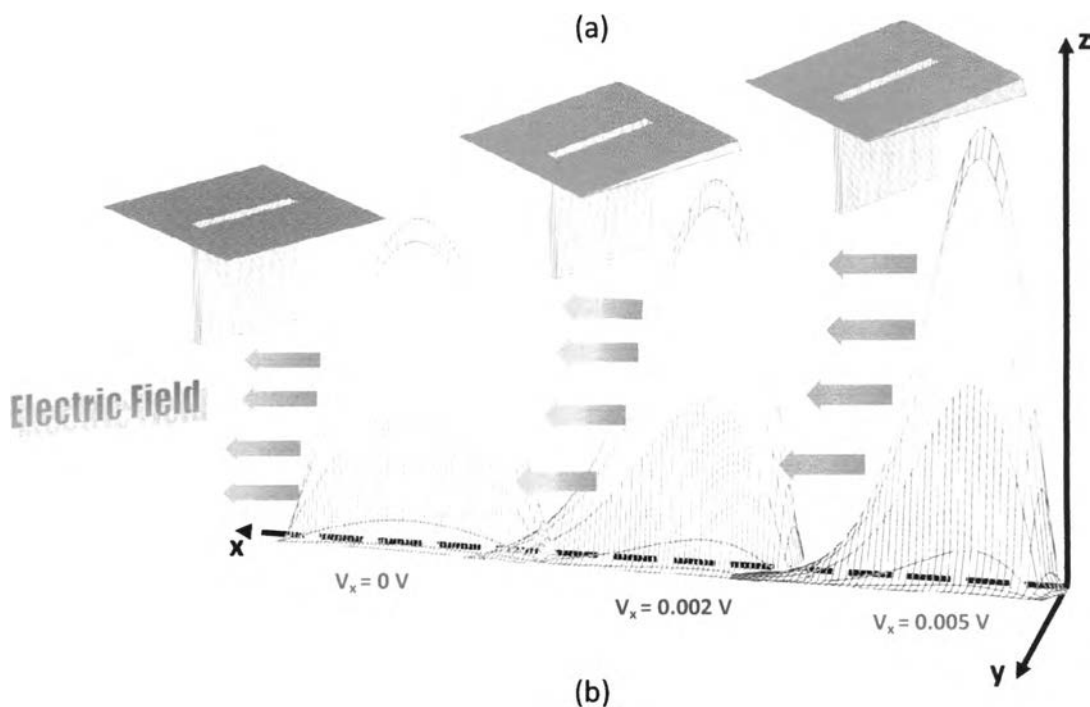
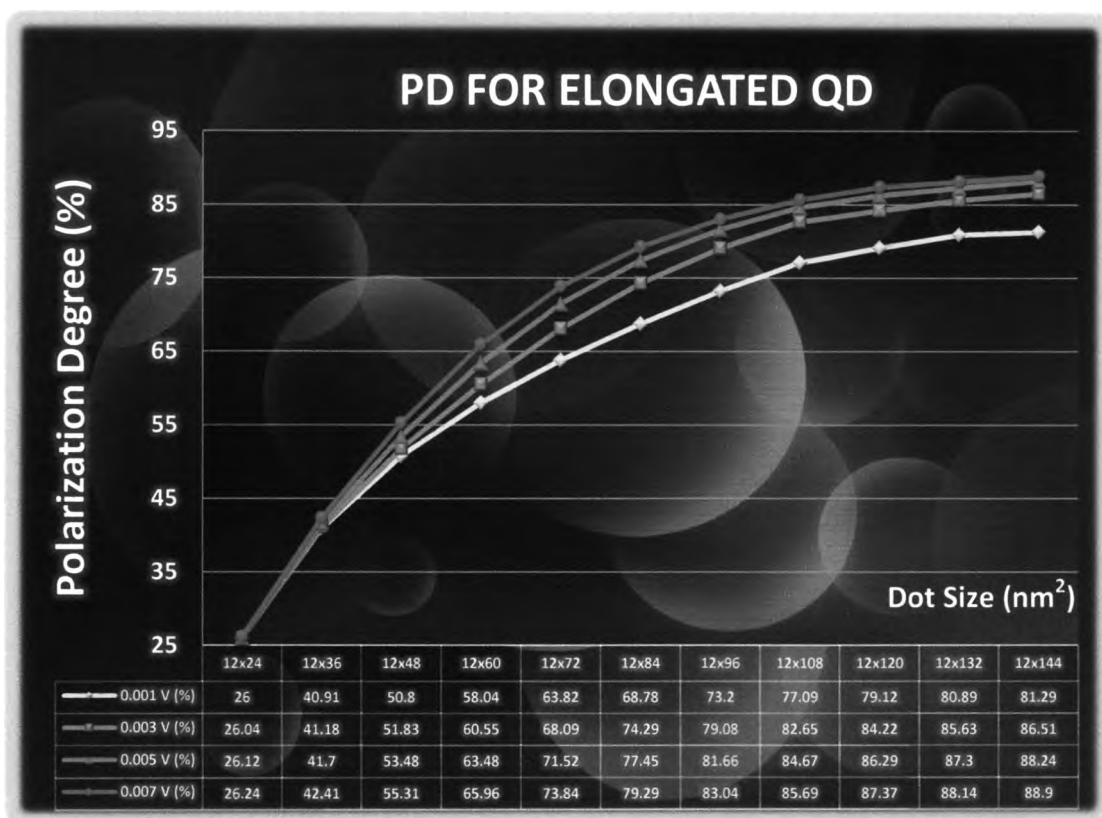
Although the electron (hole) wavefunction is changed by the electric field in the x direction, this wavefunction is oppositely attracted by hole (electron) via electron–hole Coulomb interaction. Coulomb interaction leads to a concentration of electron and hole at the center of QD (in the normal state). Because of this Coulomb interaction, the shapes of the wavefunctions in the x direction remain nearly spatially symmetric with respect to the maximum points of the wavefunctions. If there is no Coulomb interaction, the shapes of the wavefunctions become highly asymmetric in the x direction. It may be expected that applying a sufficiently strong electric field may destroy the exciton binding energy. This point will be demonstrated again in the next section. All of these results reveal that the single QD structure even with an isotropic shape may exhibit a polarized characteristic by the effect of electric field, and this agrees well with the available calculated data reported in [197].

#### 4.2.2 Optical Polarization Characteristic of Elongated QD Structure under Electric Field along the Direction of Elongation

Next, the polarization degree of asymmetric QD structure is demonstrated in the applied electric field situation. As discussed in **section 3.1.4.2.1**, only its elongated structure can provide an optical anisotropy since the length in the x (elongated) direction is larger than the y direction. The distribution function of electron and hole mainly concentrate along the x direction which in turn gives a strong lateral confinement of the carriers in the x direction, that is, a high transition probability in the x direction. These effects, on the other hand, result in the occurrence of non-zero polarization degree of the elongated QD structure.

It is interesting to know what happens when the electric field is applied parallel to the direct of QD elongation. Not surprisingly, this yields an increase of polarization degree which is greater than in case of isotropic QD. This can be described as follows. In case of isotropic QD, it is necessary to apply a sufficiently large electric field to force the electron and hole wavefunctions to become certainly localized in one direction that eliminates the symmetric distribution in order to give the polarization anisotropy. While the presence of applied electric field in the elongated structure not only accelerates the carrier movement without threshold voltage needed to initiate the transition probability in one direction to be dominant than that in the other direction, but also encourages the higher value for degree of polarization, as well (see **Figure 4.7 (b)** that the hole wavefunction distributes more in the x direction than in the case of isotropic QD and distorts more when increasing the applied voltage).

**Figure 4.7 (a)** gives a detail of PD value on the elongated QD under the applied electric field, whose size in the y direction was fixed to 12 nm and that in the x direction varied from 24 nm to 144 nm. The increase of polarization degree can



**Figure 4.7** (a) The PD of single QD under the applied voltage  $V_x$  chosen for 4 values (0.001, 0.003, 0.005, and 0.007 V) vs. the increase of QD size in the x direction elongated from 12 to 144 nm. (b) The ground-state hole wavefunction of 12x120 nm<sup>2</sup> QD under  $V_x$  of 0, 0.002, and 0.005 V, respectively.

be obviously seen during the first period of enlargement the QD size in the x direction and continually goes up until it reaches saturation point when the size in the x direction is much larger than the y direction. Therefore, the QD with a larger lateral size is necessary for increasing the carrier confinement in the x direction, which gives rise to the higher polarization degree. For all elongated QDs, the electrons and holes are separated along the direction of elongation and spatial expansion of the hole wavefunction is smaller than that of the electron wavefunction. These spatial expansions strongly depend on the extent of size elongation [197].

In the environment of electric field applied to the QD system, as the applied voltage changes from 0 V to 0.001 V, the degree of polarization of each elongated QD size also increases but not much compared with the size effect. The increase of polarization degree can be mostly seen at  $V_x$  increasing from 0.001 V to 0.003 V for the QD size of 12x144 nm<sup>2</sup> (approximately 5 % increase). The overlarge lateral sizes and the strong electric field give a similar trend, that is, their effects cause PD approaching to the constant values. In other words, this electric-field-induced optical anisotropy increases with increasing electric field and/or lateral size of QD. The benefit of applied field in this case is to gradually fine-tune the carrier transition probability to be distributed more in one direction in order to obtain a higher degree of polarization anisotropy.

#### **4.2.3 Comparison of Polarization Degree on Various QD Aspect Ratios in the Environment of Applied Electric Field**

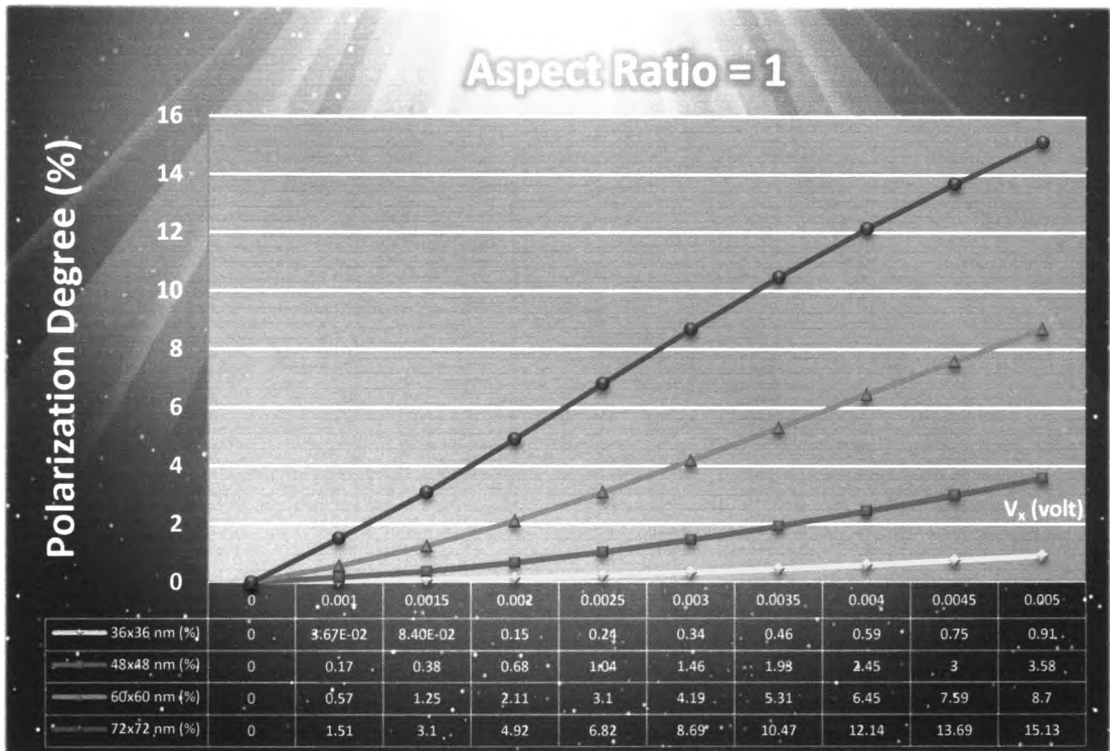
As the last case for calculating the polarization degree of single QD under the applied electric field, we consider the size effect on the QD in various aspect ratios. **Figure 4.8 (a)** illustrates the polarization degree of different isotropic QD (aspect ratio= 1) versus applied voltage  $V_x$  varied from 0 to 0.005 V. In smaller QDs, there are electrons and holes distributed nearly all over the QDs and the spatial shapes of



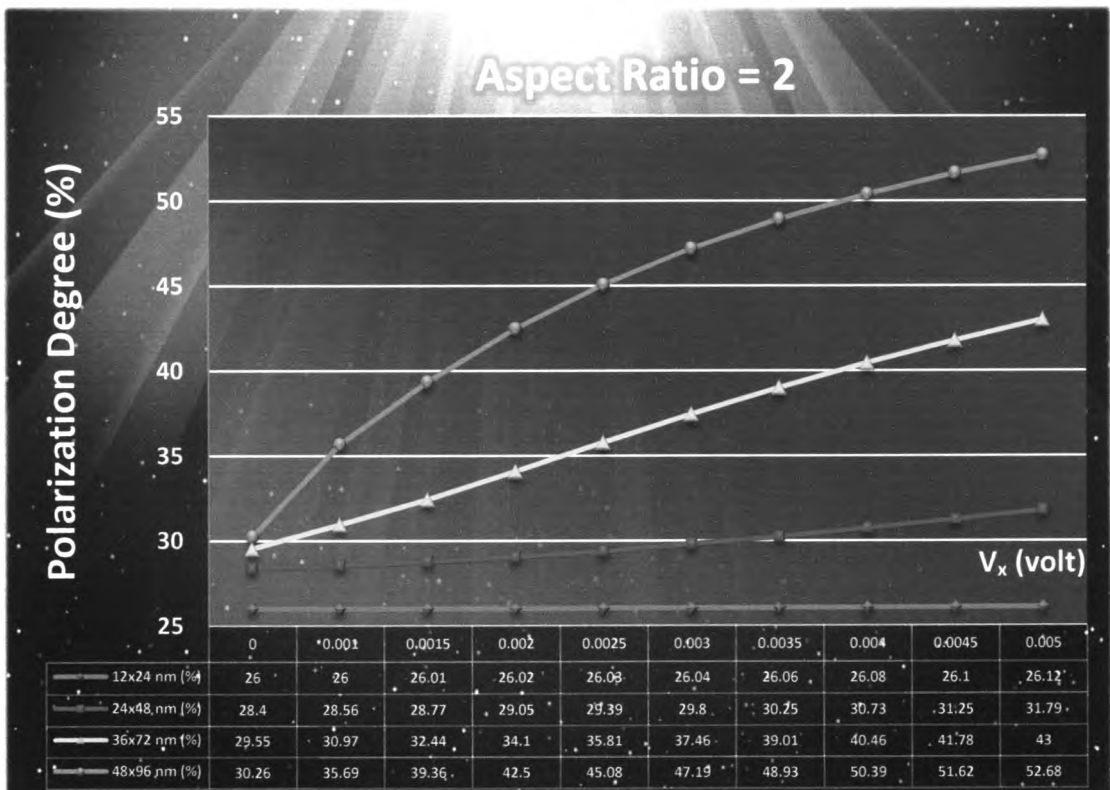
the wavefunctions are almost isotropic (the electric field does not significantly change the distribution functions of the electron and hole), leading to a small degree of polarization. Note that the widths of the electron and hole wavefunctions increases with decreasing the QD size, due to the fact that the kinetic energies of the electron and hole are larger for the smaller QD because of the stronger confinement, which enables the electron and hole to move farther away from the center of the QD. Conversely, the distribution functions of both electron and hole become narrower as the QD size increases [146].

In larger QDs, the electron and hole wavefunctions are spatially localized so this effect is prominent and does not happen in smaller dots. For large QDs, the electron and hole wavefunctions can greatly change inside the dots, and since the Coulomb interaction is small, the Stark shift is much larger for this case [65]. Thus, the wavefunctions become spatially asymmetric, and this asymmetry leads to the non-zero degree of polarization. However, this PD value remains quite low because of its isotropic shape except that the QD is an overlarge size (the 72x72 nm<sup>2</sup> QD shows a PD value about 15.13% at strong electric field) for an appreciable degree of polarization. However, the overlarge size of QD may enter to the macroscopic size which goes out of the quantum mechanical regime.

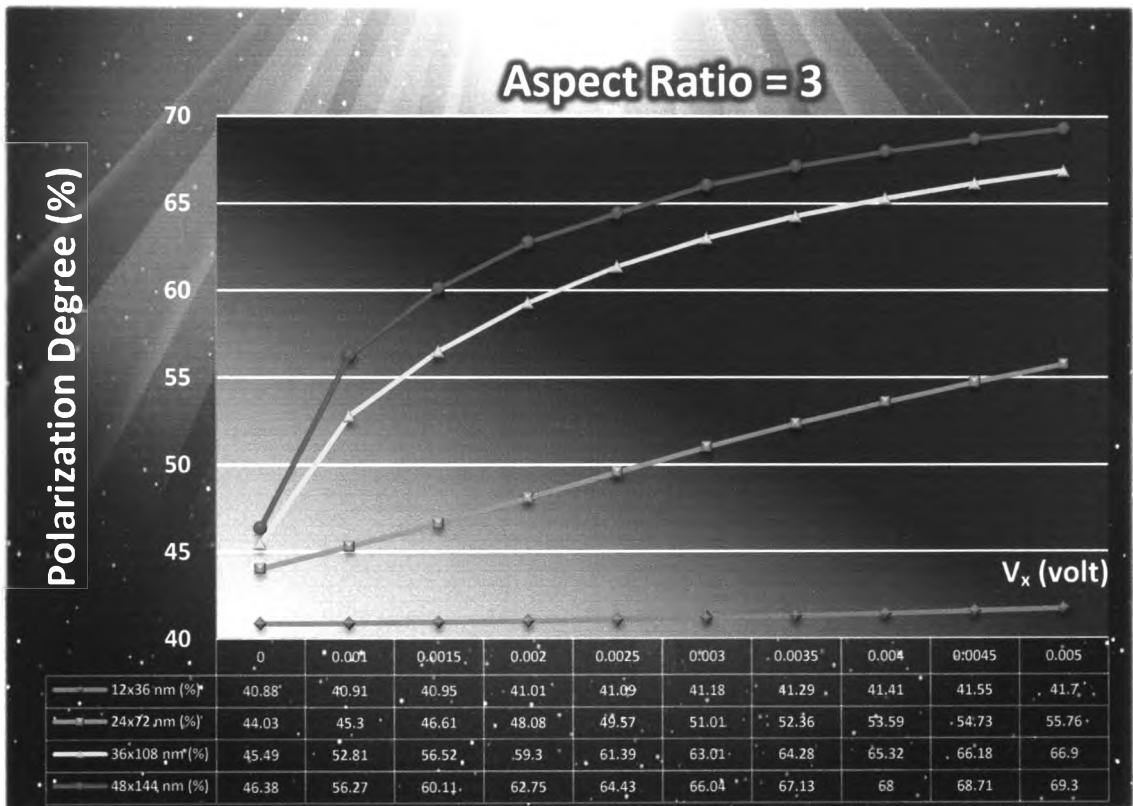
Our calculations imply the another factor related to the QCSE, that is, the average distance between the electron and hole in the x (elongated) direction or the “induced electric dipole moment” of the exciton, increases with increasing lateral electric field. The major contribution is due to the stronger lateral confinement of the hole [134, 146]. The physical meaning of the pronounced lateral field dependence of the dipole moment may be understood as follows. The hole wavefunction basically moves along the surface of QD when a lateral field is applied. The electron wavefunction, on the other hand, is more extended and remains delocalized within the entire QD up to fairly high values of the lateral field. If the top part of the QD is flat, the center of mass of the hole, then moves along the surface with increasing lateral field. This effect alone would lead to a constant dipole moment. In addition, the Coulomb interaction between the electron and hole



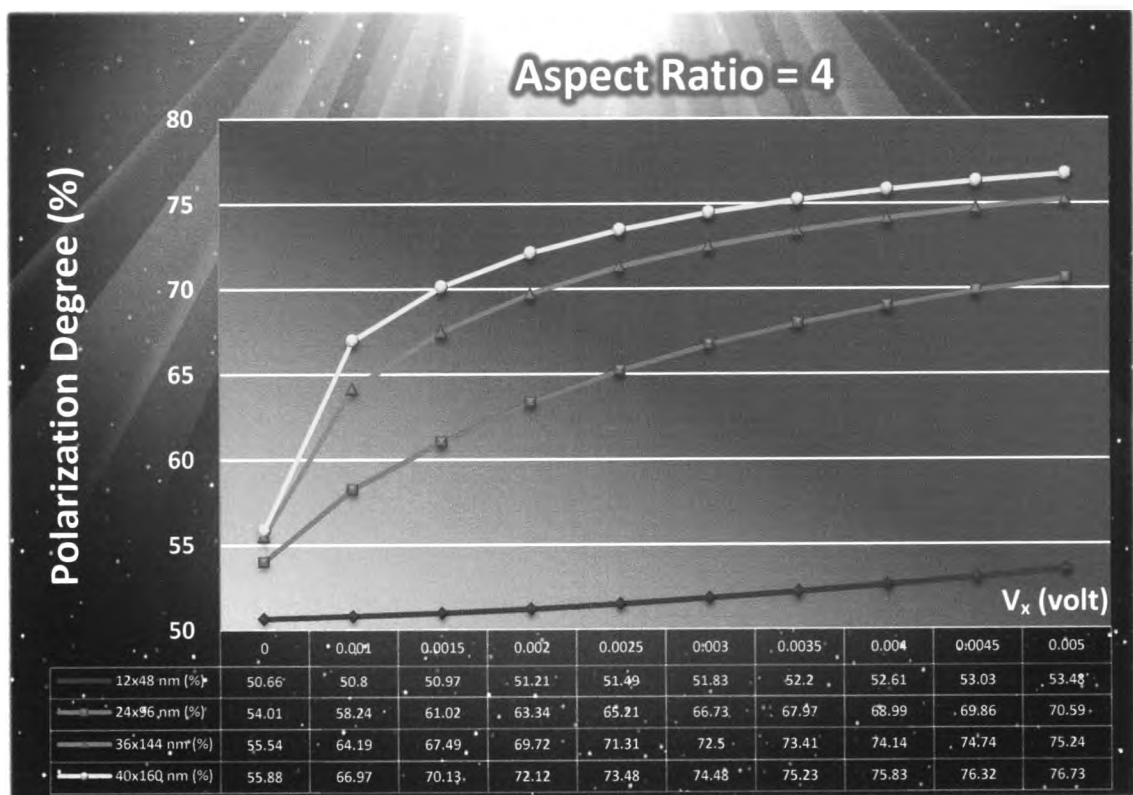
(a)



(b)



(c)



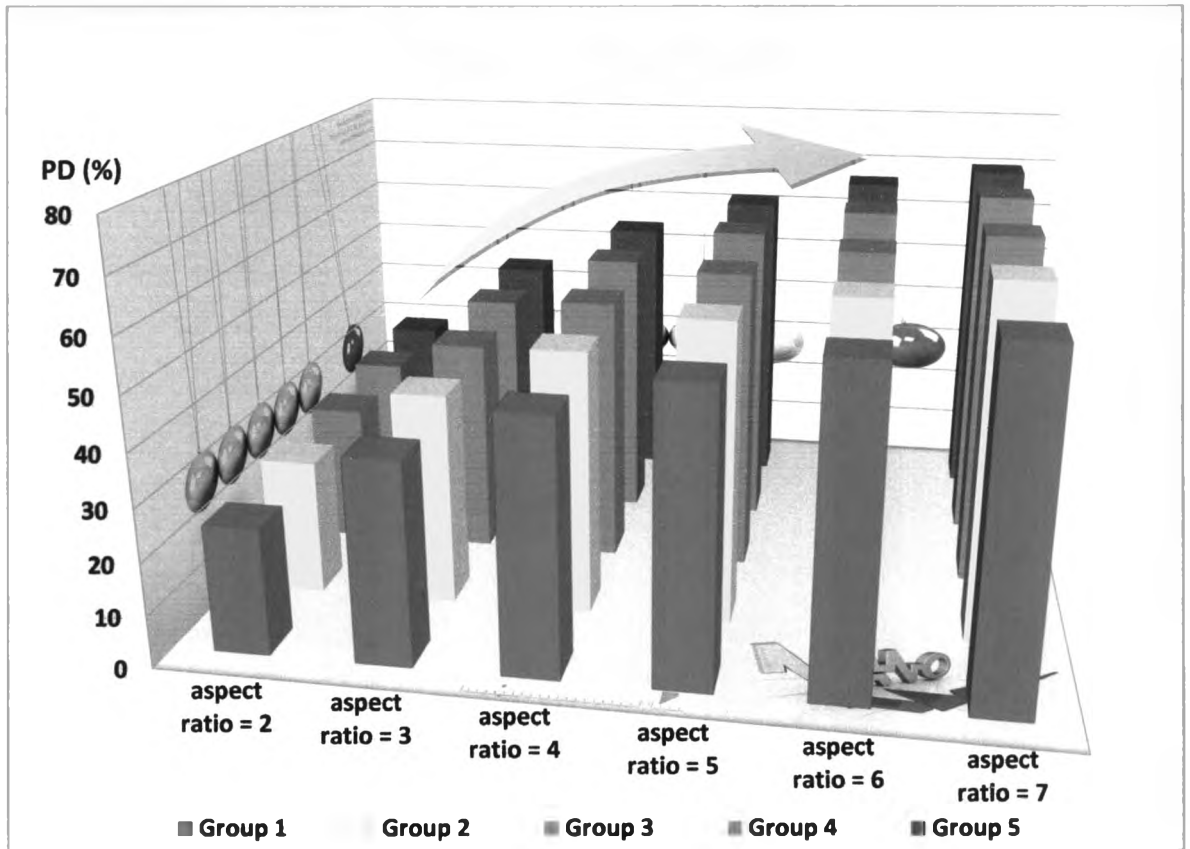
(d)

**Figure 4.8** The polarization degree of single QDs vs. applied voltage  $V_x$  (varied from 0 to 0.005 V) with the aspect ratio of (a) 1, (b) 2, (c) 3, and (d) 4, respectively.

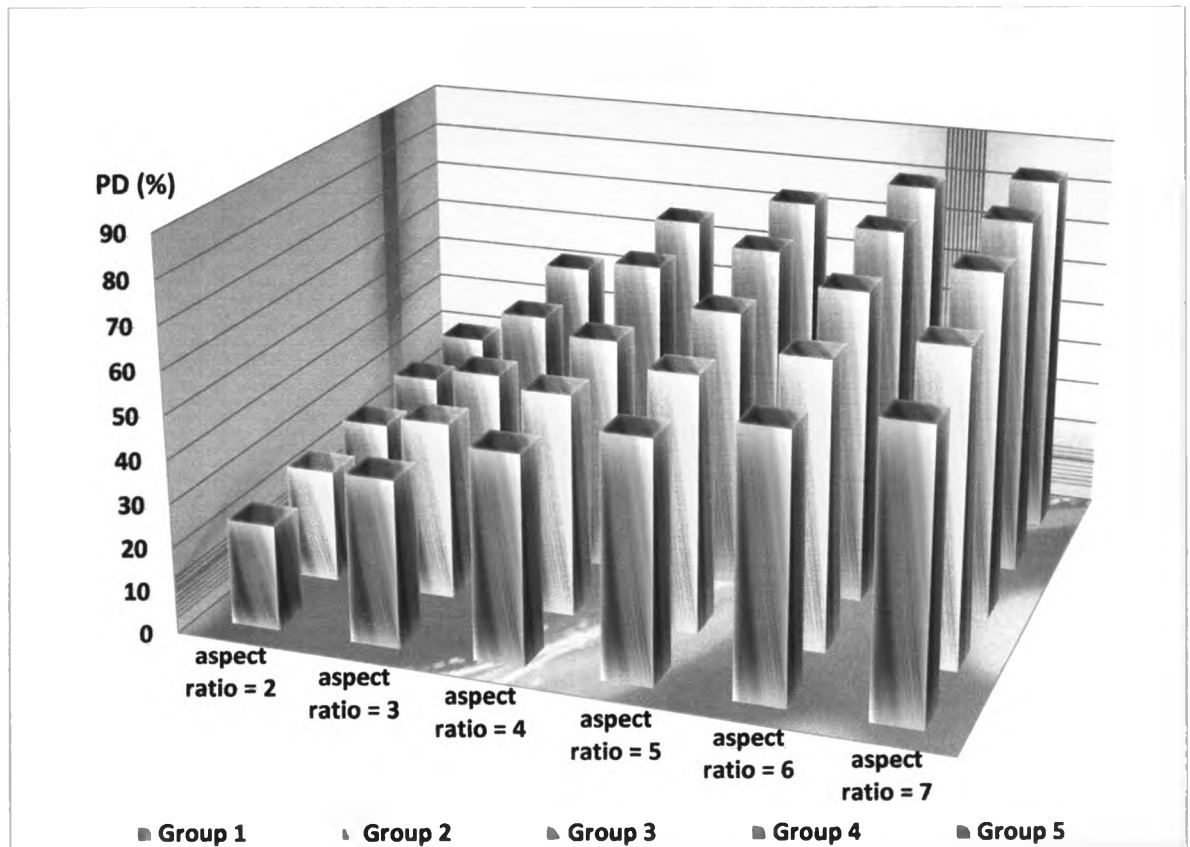
competes with the QCSE in determining the induced electric dipole moment of the exciton. In fact, the effect of induced electric dipole moment, which is closely related to exciton, also significantly depends on the shape of QD and its alloy profile for a given lateral electric field (we will not delve much into the detail because this is out of scope of the thesis work).

The other aspect ratios of QD size (2, 3, and 4, as shown in **Figure 4.8 (b), (c), and (d)**, respectively) including additional data on the other polarization degree of various sizes, aspect ratios, and applied voltages (**Figure 4.9**) were plotted to examine the PD values (Note that all charts in **Figure 4.9** were called “Manhattan bar chart” [230]. The advantage of this chart type is to display series of individual bars grouped by category in three-dimensional view that is appropriate for a lot of data, attractiveness, and easy to understand way). All of the results show that their tendency are in the same way due to their size non-uniformity (or elongated structures), and consistent with the calculations demonstrated in **section 4.2.2** (the preliminary explanation was already described in **section 3.1.4.2.1**).

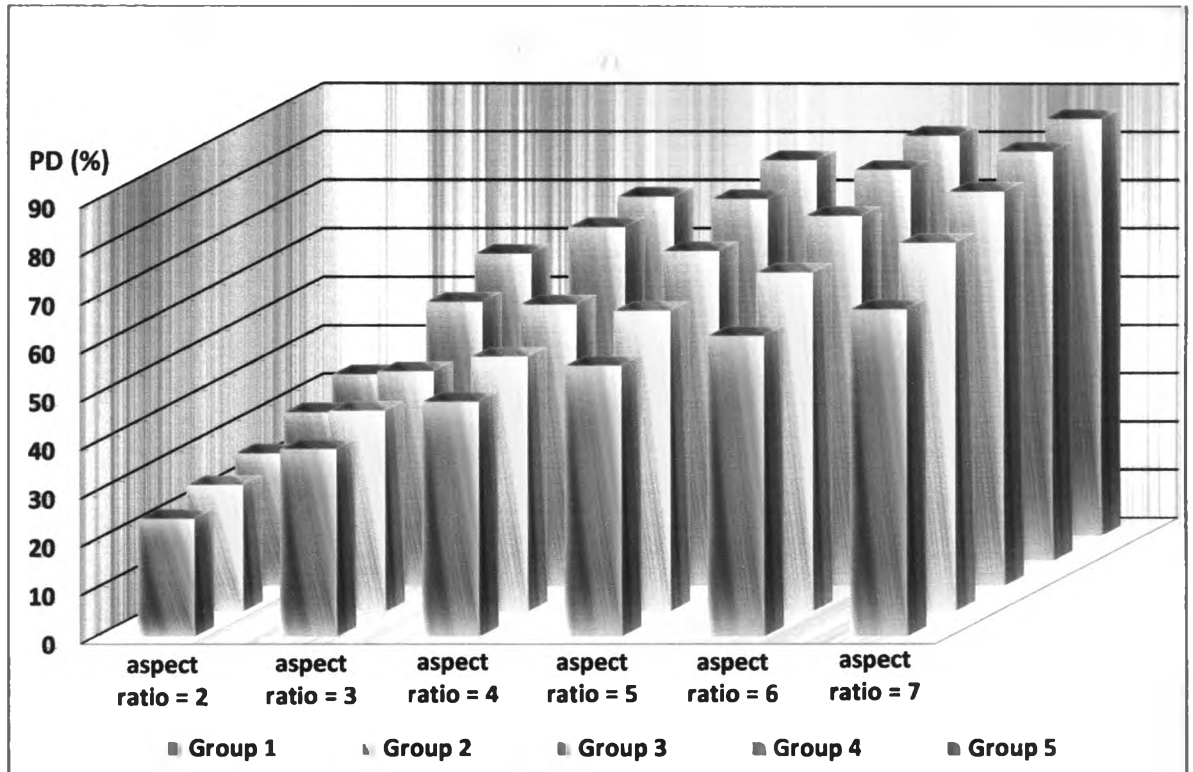
As the voltage is applied to the system, we see that the smaller dot sizes are less affected by the electric field than in the larger dots displayed by the curvature of PD values in case of large QDs being larger than the smaller QDs. Despite increasing the dot size only in the one direction shows a high PD value (because of the elongated structure that strongly confined the carriers, indicated as a high aspect ratio), the effect of size extended in both directions and electric field strength sometimes may draw a comparison with it, represented by the examples as follows:



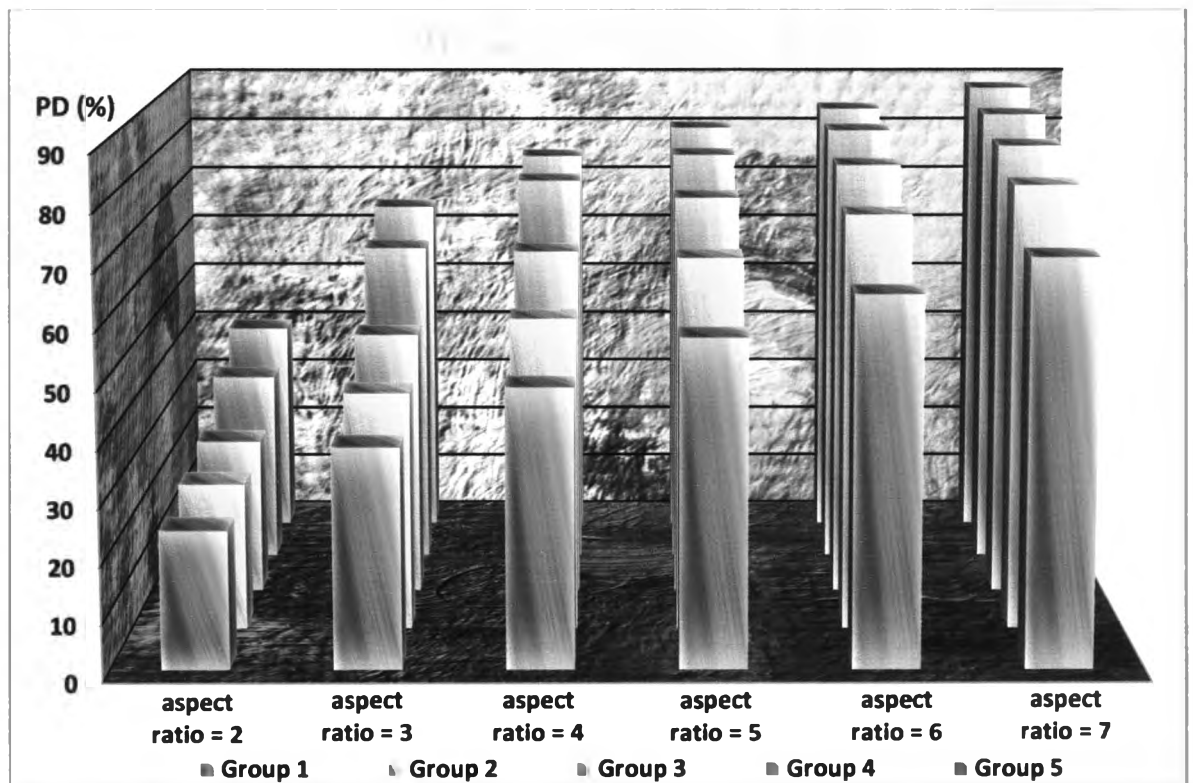
(a)



(b)



(c)



(d)

**Figure 4.9** Manhattan bar chart for PD values of QDs with various sizes and aspect ratios under the applied voltage of (a), 0 (zero field) (b), 0.002 (c), 0.004 and (d) 0.008 V, respectively.

**Table 4.1:** Comparison of PD values with various sizes, aspect ratios, and applied voltages.

Aspect Ratio	Group 1		Group 2		Group 3		Group 4		Group 5		
	size (nm <sup>2</sup> )	PD (%)	size (nm <sup>2</sup> )	PD (%)	size (nm <sup>2</sup> )	PD (%)	size (nm <sup>2</sup> )	PD (%)	size (nm <sup>2</sup> )	PD (%)	
V <sub>0</sub> = 0.002 V	2	8x16	24.28	12x24	26.02	16x32	27.14	24x48	29.05	28x56	30.28
	3	8x24	38.61	12x36	41.01	16x48	42.88	24x72	48.08	28x84	51.78
	4	8x32	48.26	12x48	51.21	16x64	54.38	24x96	63.34	28x112	66.35
	5	8x40	55.32	12x60	59.13	16x80	64.05	24x120	70.01	28x140	73.63
	6	8x48	60.86	12x72	65.92	16x96	71.97	24x144	77.43	28x168	80.16
	7	8x56	65.48	12x84	71.83	16x112	79.14	24x168	82.3	28x196	84.02
	V <sub>0</sub> = 0.004 V	2	8x16	24.29	12x24	26.08	16x32	27.39	24x48	30.73	28x56
3		8x24	38.66	12x36	41.41	16x48	44.41	24x72	53.59	28x84	58.31
4		8x32	48.47	12x48	52.61	16x64	58.2	24x96	68.99	28x112	70.11
5		8x40	55.89	12x60	62.05	16x80	68.2	24x120	74.8	28x140	77.64
6		8x48	62.07	12x72	69.97	16x96	76.49	24x144	80.84	28x168	82.75
7		8x56	67.53	12x84	76.1	16x112	81.52	24x168	94.64	28x196	86.24
V <sub>0</sub> = 0.008 V		2	8x16	24.33	12x24	26.31	16x32	28.29	24x48	35.18	28x56
	3	8x24	38.89	12x36	42.82	16x48	48.99	24x72	60.31	28x84	64.45
	4	8x32	49.25	12x48	56.21	16x64	63.96	24x96	73.41	28x112	74.68
	5	8x40	57.78	12x60	67.77	16x80	73.91	24x120	78.37	28x140	86.33
	6	8x48	65.16	12x72	74.72	16x96	79.96	24x144	83.1	28x168	84.14
	7	8x56	71.45	12x84	79.95	16x112	83.54	24x168	86.14	28x196	88.16

- The PD value of  $12 \times 144 \text{ nm}^2$  QD at  $V_x = 0.007 \text{ V}$  is 88.9% (see **Figure 4.7 (a)**) which is rather high due to very elongated structure (aspect ratio = 12), but the PD in case of  $28 \times 196 \text{ nm}^2$  QD also manifests the high value about 86.24% (see **Table 4.1**) which is slightly different than the former case (< 3%) even though it has a smaller aspect ratio (7) and applied voltage (0.004 V). (The effect of size-extension in all directions overcomes the lateral size-extension effect.)

- The PD value of  $16 \times 64 \text{ nm}^2$  (aspect ratio = 4) at  $V_x = 0.008 \text{ V}$  is 63.96% (see **Table 4.1**) is approximate to the PD value of  $12 \times 72 \text{ nm}^2$  (aspect ratio = 6) at  $V_x = 0.001 \text{ V}$  which is 63.82% (see **Figure 4.7 (a)**) though it has a smaller aspect ratio due to the assistance of stronger voltage.

These calculated data indicate that in case of the same aspect ratio, the larger the QD size, the higher the PD. Besides, at the large aspect ratio, PD value also increases (the more elongated structure, the larger PD value is obtained). Nevertheless, it is essential to compromise between the size effect and the electric field strength by properly keeping the aspect ratio of both directions (not overlarge since its structure is impossible to grow successfully in practice) with a suitable electric field for given a strong polarization anisotropy.

Again, an optical anisotropy is clearly prominent in larger QDs under a “strong” electric field because the hole wavefunction is largely localized and high polarization degree appears (their polarization properties directly reflect the spatial symmetry of the hole wavefunction, as described in **section 4.1.1**). However, in smaller QDs, large “optical responses” are achieved due to the large Coulomb interaction [197], by contrast, the optical responses become weaker in the larger QDs when a “strong” electric field is applied. In larger QDs, the spatial symmetry of hole wavefunction greatly changes, causing a large optical anisotropy.



Simultaneously, the electrons and holes are widely separated from one another inside the quantum dot, leading to a small spatial overlap between the electron and the hole wavefunctions. This results in lower oscillator strength and weaker optical response. These explanations also insist on the effect of relative recombination rate [146]. In larger QDs, the electric field dependence of the exciton recombination rate is much stronger compared with that of smaller QDs. Its exciton recombination rate goes down slowly initially at the low field, decreases very fast when the applied field is “very large” in the order of 0.01 - 0.02 V/nm (the exciton lifetime in this case is about six times that under zero field), and slowly saturates again when the field strength is above 0.02 V/nm. The results are consistent with the fact that a larger QDs is more influenced by the electric field due to a weaker confinement. Furthermore, decreasing of recombination rate (considered only radiative recombination) of larger QDs under “strong” applied field suggests the reduction of optical responses as well (larger radiative lifetime).

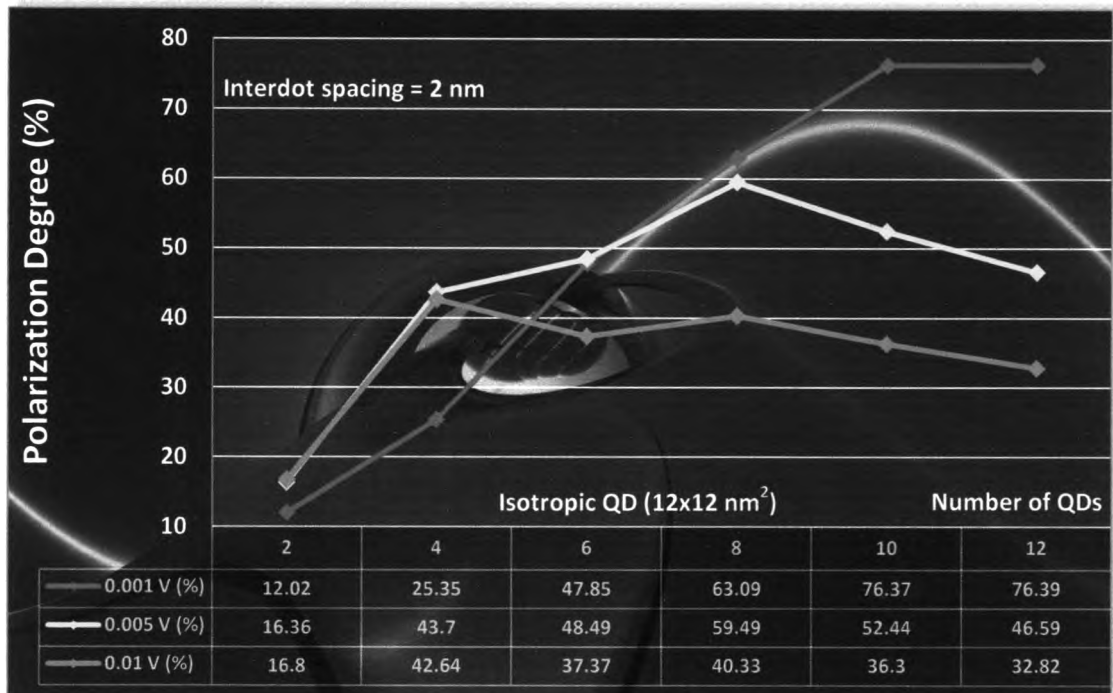
The optical polarization study of single QDs is completely investigated. Hereafter, we will pay attention to the ordered QD case which is the main point of this thesis. The calculation results and physical explanation were performed to know the exact information of linearly aligned QDs and compare with the all previous cases to draw an entirely conclusion of thesis work.

### 4.3 LPD CHARACTERISTIC OF ALIGNED QDS UNDER THE ELECTRIC FIELD

At this moment, investigation of optical properties on QDs under the applied electric field has reached at a main part of the thesis work. Studying should be extended to a more complicated structure, i. e., linearly aligned QDs. It involves additional number of QDs, particularly in the occurrence of “overlap integral” which is one of the important factors because this parameter changes rapidly with spacing between QDs, and then is used to determine degree of polarization of the QD system (as discussed in **section 3.1.4.2.2**). Therefore, we anticipate an interesting variation when electric field is applied related to the effects such as, interdot coupling and wavefunctions distribution, Coulomb interaction and dipole moment, quantum Stark, and tunneling effect (electroabsorption), etc. These are also given an explication that supports the experimental results providing a clarification in physical meaning to understand these mechanisms logically. At last, the total researching information will be systematized again for contemplation in the end of line to sum up an overall discussion in the last section of this chapter.

#### 4.3.1 Effects of Electric Field Strength on the Polarization Degree

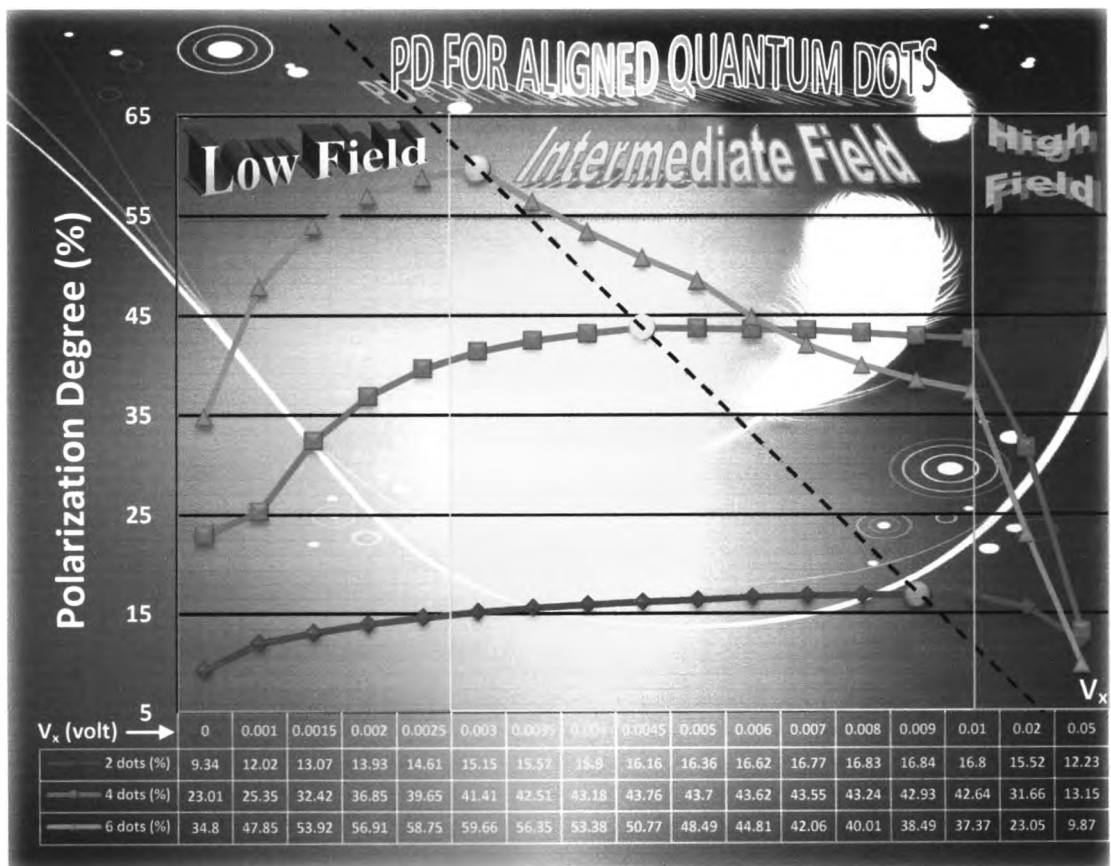
We first check the PD's tendency roughly by plotting the polarization degree of isotropic shape QDs ( $12 \times 12 \text{ nm}^2$ ) with interdot spacing of 2 nm versus the number of QDs at three different constant voltages of 0.001, 0.005, and 0.01 V, respectively. The result is schematically shown in **Figure 4.10**. It is seen that for the applied voltage of 0.001 V, the polarization degree increases rapidly as the number of QDs increases until slowly increasing when reaches to the twelve dots, so we expect to show a same nice results for the other two cases. Unfortunately, the polarization degree becomes fluctuate, the results is therefore unpredictable. Our presumption believe that the electric field strength may be strongly affects the PD value and closely related to variation of the overlap integral, thus we need to probe this thoroughly in the view of various range of applied voltage henceforth.



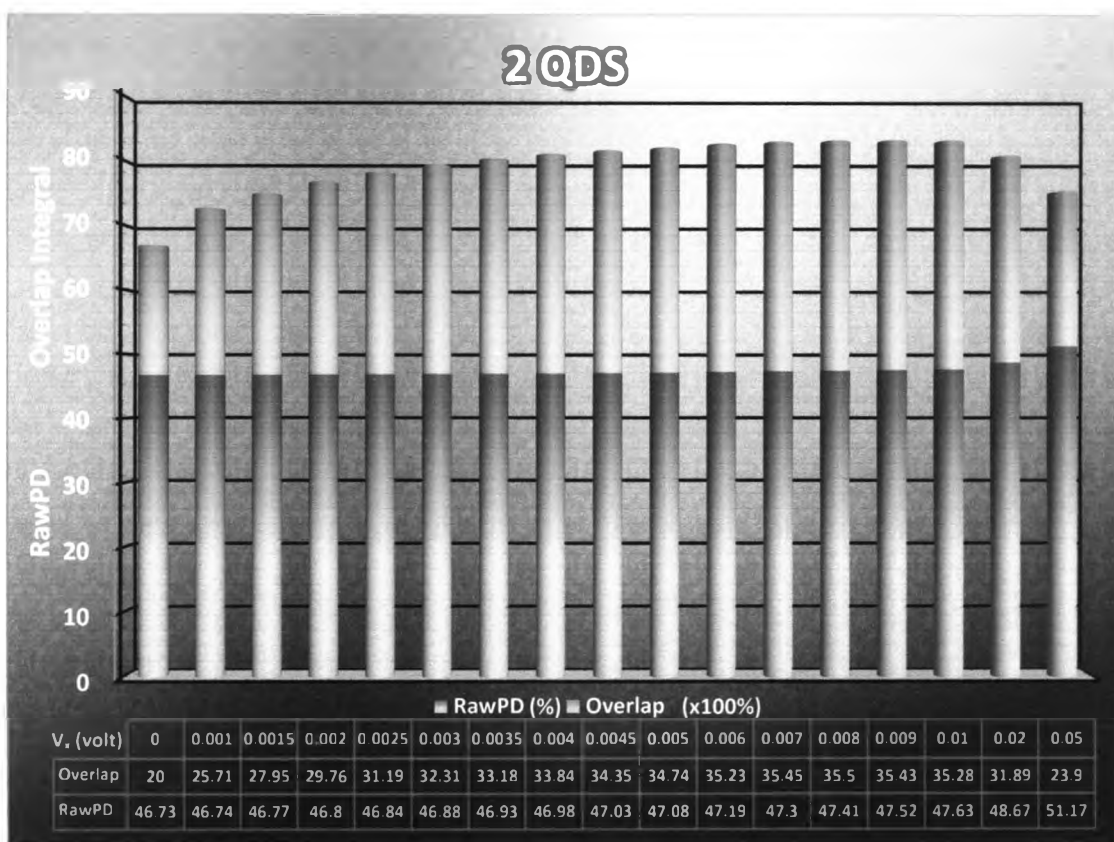
**Figure 4.10** The polarization degree vs. the number of 12x12 nm<sup>2</sup> QDs with their spacing of 2 nm under the applied voltage fixed at 0.001, 0.005, and 0.01 V, respectively.

**Figure 4.11 (a)** shows the polarization degree of three QDs groups: 2, 4, and 6 dots whose size and interdot spacing were the same as the previous case mentioned above. The voltage applied to the system was varied from 0 to 0.05 V. Note that it is the most remarkable graph in the thesis work, so it is specially highlighted since it gives several phenomena which are connected to important aspects, both main points and several subpoints. Focusing on the result, it is observed that PD values of all lines from three cases are separated into three regions, corresponding to the electric field strength as follows: low field (pink region), intermediate field (yellow region), and high field (cyan region), respectively. For binary QDs (blue line), the PD value slowly goes up following the increase of voltage from 9.34% (zero voltage) to the maximum value of 16.84% (pink circle) at voltage of 0.009 V (represented as  $V_{PD(max)}$  or of voltage producing the PD maximum) before it slowly decreases to 12.23% at voltage of 0.05 V which is in the strong field region. In case of four QDs (red line), the PD value at low field also shows the same trend as for two QDs, which

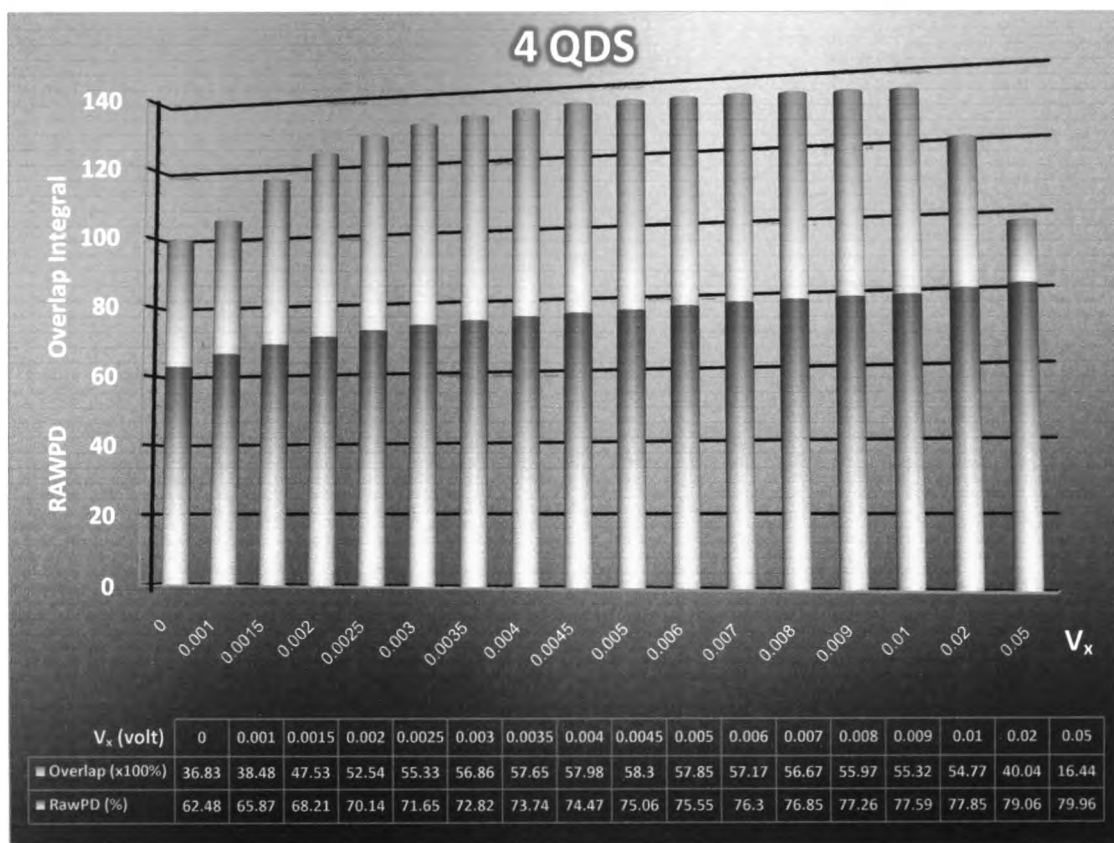
more rapidly increases until it reaches the maximum value of 43.76% (yellow circle) occurring at lower  $V_{PD(max)}$  (0.0045 V or half value compared to the binary QD case). After that, it slowly decreases but applied voltage is still in the range of intermediate field region. Unexpectedly, when the voltage is larger than 0.01 V or entering to the high field region, the PD value sharply decreases, and decreases further and further to 13.15% as the maximum voltage, which is lower than in the case of no applied voltage (23.01%). The variation of polarization degree of six QDs (green line) is similar to the four QD case but the decrease of the PD value in the intermediate and high field regions can be more obviously seen by the PD maximum value (orange circle) reached at  $V_{PD(max)} = 0.003$  V (the difference  $\Delta V_{PD(max)}$  compared to the four QD case ( $\Delta V_{PD(max)} = 0.0045 - 0.003 = 0.0015$  V) is less than when compared to the former case ( $\Delta V_{PD(max)} = 0.0045$  V)), and PD at the maximum voltage used turns out to be lower than the binary QD case (9.87%). From the results mentioned above, we see that PD of these cases rapidly decreases in the strong applied voltage and decreases even more when increasing the number of QDs which confirms that the electric field strength strongly affects the PD, consistent with previous hypothesis.



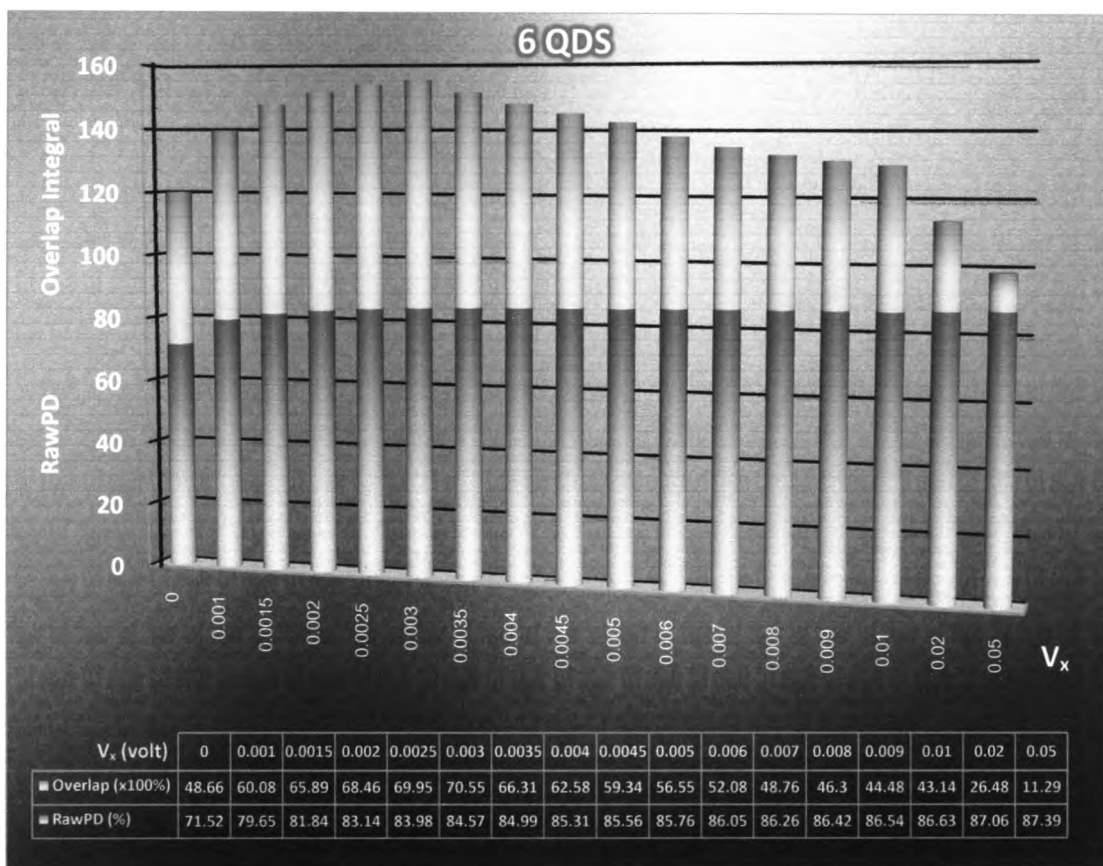
(a)



(b)



(c)



(d)

**Figure 4.11** (a) The polarization degree of 2, 4, and 6 QDs and the stacked bar chart of RawPD and overlap integral (x100%) values corresponding to (b) 2, (c) 4, and (d) 6 dots, respectively under the applied voltage varied from 0 to 0.05 V. The QD size and distance between them are the same as **Figure 4.10**.

**Figure 4.11 (b), (c), and (d)** display the RawPD and the overlap integral ( $\times 100\%$ ) values in form of stacked bar chart utilized for showing the relationship of parts to all the variables of interest (in this place, we focus on total polarization degree which is composed of the RawPD multiplied with the overlap integral). The results show that the RawPD of these cases gradually monotonically increase with increasing applied voltage. By contrast, the tendency of overlap integral in all cases are similar to the variation of PD values under the voltage applied to the system, which implies that the total PD is strongly influenced by the overlap integral. Hence, the carrier interaction plays a crucial role in determining the overlap integral, as well. The physical explanation according to all the above results is elucidated as follows:

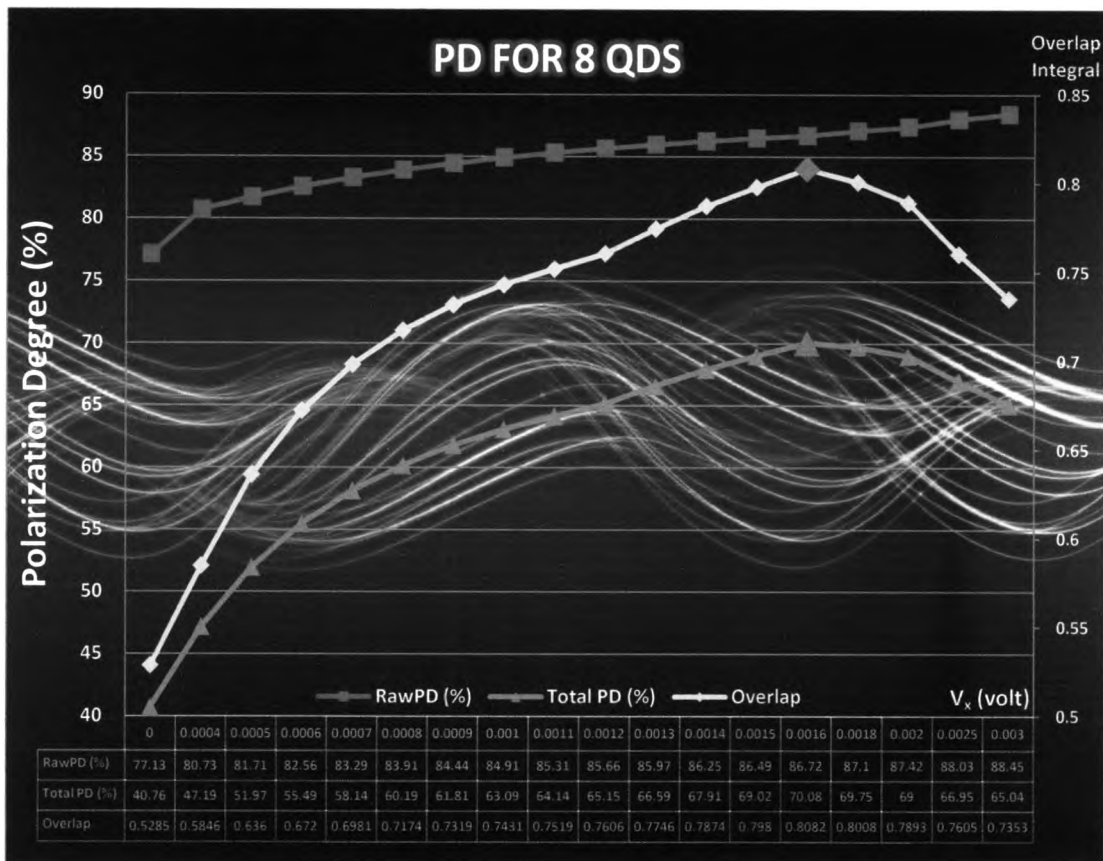
- In the low applied electric field region (0 - 0.001 V/nm), the Stark effect is weak and the electron and hole behave more like a pair which induces a small effective dipole moment. Nevertheless, this QCSE enables both electron and hole wavefunctions to be slightly distributed in the opposite directions because the Coulomb interaction of excitons is still strong enough to hold the excitons, which prevents the separation of carriers, causing them to return to their symmetric state. Regarding the overlap between carrier wavefunctions, recall that in case of single QD the overlapping of electron and hole wavefunctions inside QD decreases when the magnitude of electric field goes up. The situation is different in case of aligned QDs (because it also includes with the coupling effect between QDs). As the applied field increases, though it causes the wavefunction distortion, the electric field pushes some part of the wavefunctions (especially in their tails) to appear more outside the potential well (or barrier region between dots), leading to the wavefunctions intensified at interdot coupling region. An enhancement of this interdot-wavefunctions (both electron and hole) overlapping region is highest occurred at the endways of wavefunctions group or overlapping region between QDs pair at head of the aligned QDs (for electron (hole) wavefunctions, it manifests in the same way (opposite) of electric field's direction). From **Eq. (3.13)**, the  $\Gamma$  value thus increases resulting in the increase of polarization degree from initial state (in the absent of electric field).

- In the intermediate applied electric field state (0.001 – 0.01 V/nm), the induced electric dipole moment increases more rapidly. The grow-up intensity of electric field leads to a high probability of carriers in tunneling outside the well and move away to the other dots. Since the electric field intensity distribution is goes down when distance from the voltage source is larger, the carriers of QDS, for example, electrons that closely the voltage source are performed by a higher electric field and move with a higher acceleration (higher kinetic energy) to coming near the adjacent electrons along the electric field's direction (the effect shows an opposite direction of electric field for holes). The effect of electric field separates the electrons and holes to move far away more and more from each other and Coulomb interaction of excitons in this disturbance condition is so weaken, that is, overlap integral now is

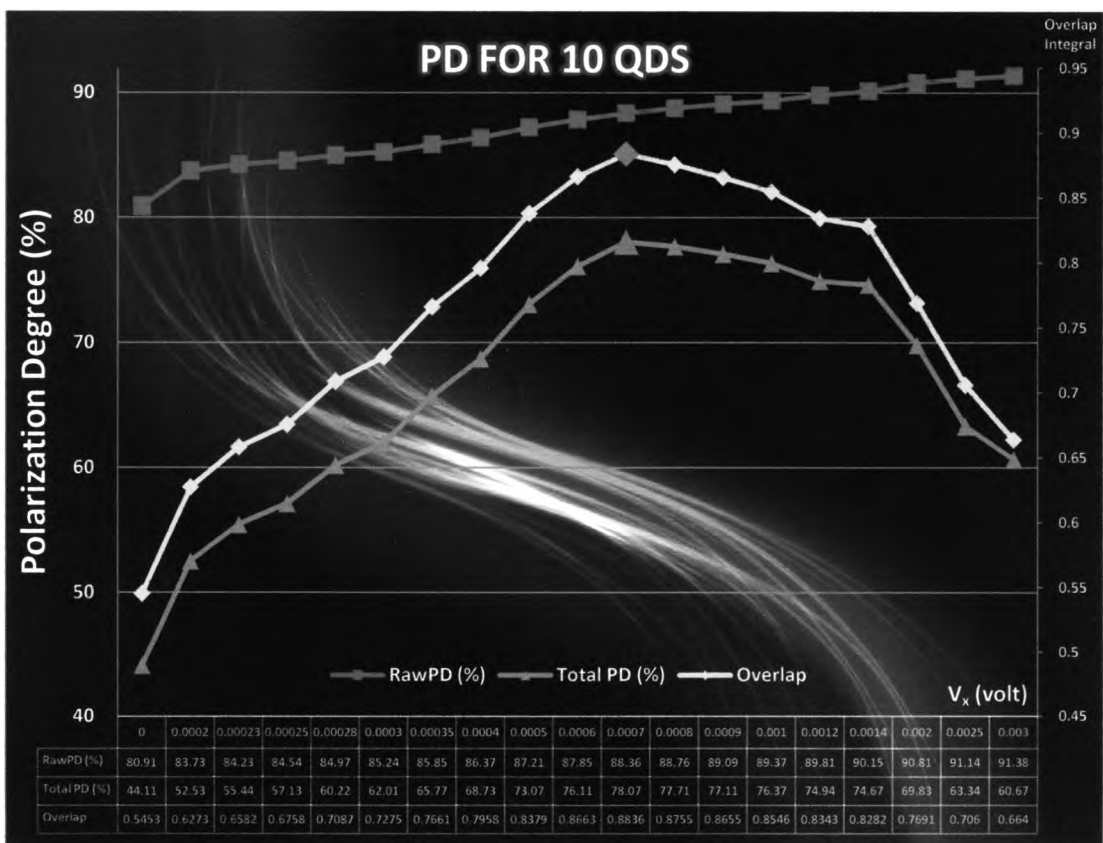
dominated by coupling interaction from applied field than the excitons Coulomb interaction. This gives a decreasing of distance between carriers which allows for stronger carriers coupling, the overlapping wavefunctions between QDs therefore increase producing a high polarization degree. Furthermore, the higher number of QDs (more overlapping region due to additional dots), the higher degree of PD value, as well (though the additional excitons produce a stronger Coulomb interaction, but this effect stills unable to compete with the coupling effect). The polarization degree continually goes up with an increasing electric field to the maximum value. This biased voltage brought the PD max or " $V_{PD(max)}$ " is represented as "operating point" of the system related to the optimized value generating the highest overlapping and gives rise to the maximum value of polarization degree which is an significant parameter for highest quantum efficiency.

Another important consequence is that there is a shift of  $V_{PD(max)}$  to lower voltage when increases the number of QDs. This extension of the Stark shift (in form of applied voltage shift) is advocated by the PD results for the higher number of QDs: 8, 10, and 12 QDs (repeating the same measurement with identical parameters by change only the magnitude of electric field), as depicted in **Figure 4.12 (a), (b), and (c)**, respectively. The increase of QDs can reduce the magnitude of applied electric field causing the increase of PD value to the range about 0 – 0.001 V (decreases approximately three - ten times compared to the previous cases) or shift the electric field response from intermediate field to the low field, indicating to less usage of voltage energy for given a maximum PD value. In addition, the higher number of QDs also reduce the threshold voltage or " $V_{threshold}$ " (the first voltage value that gives the increasing of polarization degree from the initial state (zero field)) of the aligned QD system.

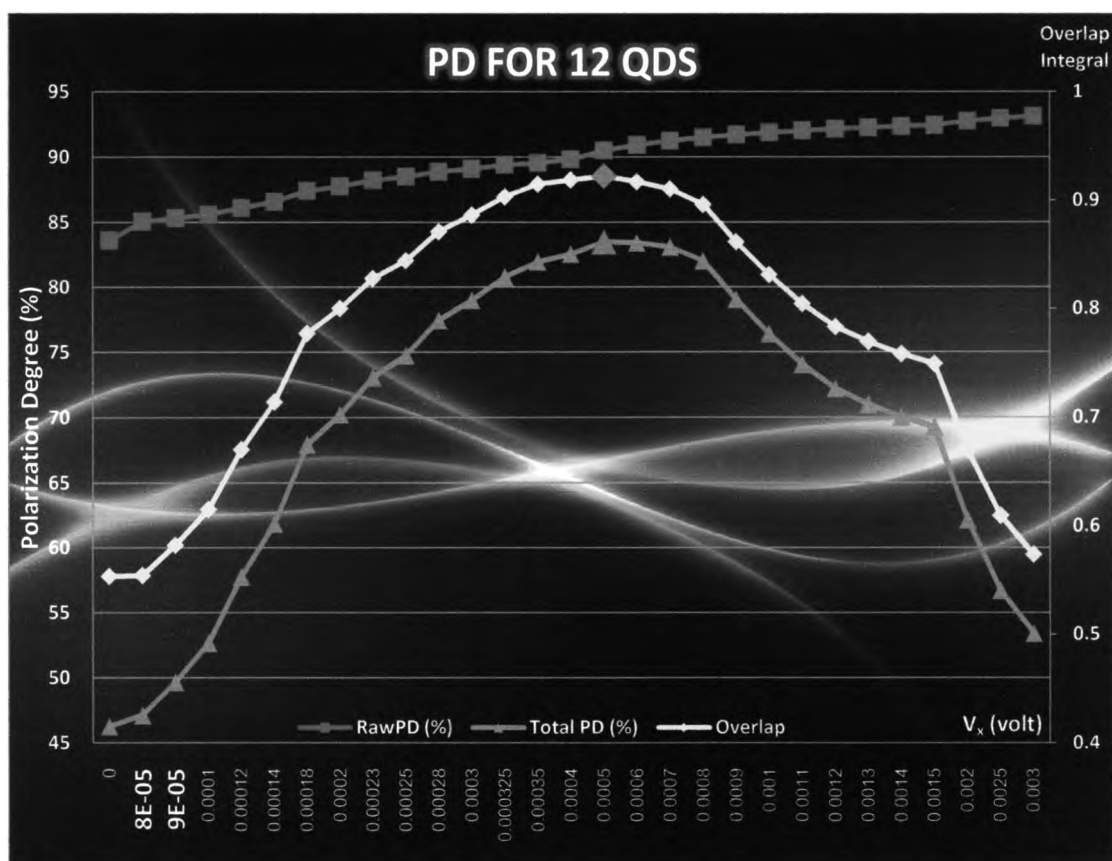




(a)



(b)



(c)

**Figure 4.12** (a) The PD values (left perpendicular axis) and overlap integral (right perpendicular axis) of 8, 10, and 12 QDs under the applied voltage varied from 0 to 0.003 V. The QD size and distance between them are the same as **Figure 4.10**.

**Table 4.2:** The polarization degree vs. applied voltage of twelve QDs.

VoltX ( $\times 10^{-3}$ V)	Overlap Integral	Raw PD (%)	Total PD (%)	VoltX ( $\times 10^{-3}$ V)	Overlap Integral	Raw PD (%)	Total PD (%)	VoltX ( $\times 10^{-3}$ V)	Overlap Integral	Raw PD (%)	Total PD (%)
0	0.5535	83.61	46.28	0.28	0.8715	88.87	77.45	1	0.8314	91.88	76.39
0.08	0.5542	85.06	47.14	0.3	0.8868	89.09	79	1.1	0.8047	92.03	74.05
0.09	0.5823	85.33	49.68	0.325	0.9036	89.4	80.78	1.2	0.7834	92.15	72.19
0.1	0.6155	85.59	52.68	0.35	0.9152	89.55	81.96	1.3	0.7696	92.26	71
0.12	0.6708	86.1	57.76	0.4	0.919	89.83	82.55	1.4	0.7583	92.36	70.03
0.14	0.7144	86.57	61.85	0.5	0.9225	90.51	83.5	1.5	0.7491	92.44	69.25
0.18	0.7775	87.4	67.95	0.6	0.9174	90.93	83.42	2	0.6701	92.76	61.26
0.2	0.8006	87.76	70.26	0.7	0.9107	91.25	83.1	2.5	0.6097	92.97	56.69
0.23	0.8282	88.23	73.07	0.8	0.8964	91.5	82.02	3	0.5735	93.13	53.41
0.25	0.8446	88.5	74.75	0.9	0.8619	91.71	79.05				

The anomaly of PD variation under applied field is happen when the voltage is greater than  $V_{PD(max)}$ , the distance between adjacent electrons (holes) is no close anymore which we believe to originate from the adjacent electrons are closed too much to make an occurrence of Coulomb force of the same charge acting against the energy force from electric field that is advantageous for excitons Coulomb interaction to pull them back. However, the electric field effect remains prominent than excitons Coulomb interaction, but the overlap integral returns to decrease slowly. This is due to the fact that when the applied field is strengthened, the band-tilting effect also increases. If this effect occurs excessively (sloped more respect to the horizontal), it leads to decreasing the height of band offset energy between GaAs capping layer and InAs QDs (the bottom of quantum well is elevated), so it is easier for carrier to move out at the top edge of quantum well, and is able to stimulate the carriers transition to become more in the y direction. In addition, a high voltage may produce a thermal energy resulting in the appearance of additional phonon energies to assist in increasing the kinetic energy and give a chance for carriers to jump out of the quantum well. Consequently, the transition probability that is formerly high in the x direction (by tunneling outside of the wall) begins decreasing, and increasing for the transition probability in the y direction at this moment ( $I_y$  also increased). Not only gradually smaller distributes of electrons and holes wavefunctions in that x direction (increase of symmetrically waveform), but also get decrease in the  $I_x$  and overlap integral. The reduced overlap integral drags into a corresponding reduction in the polarization degree of the system.

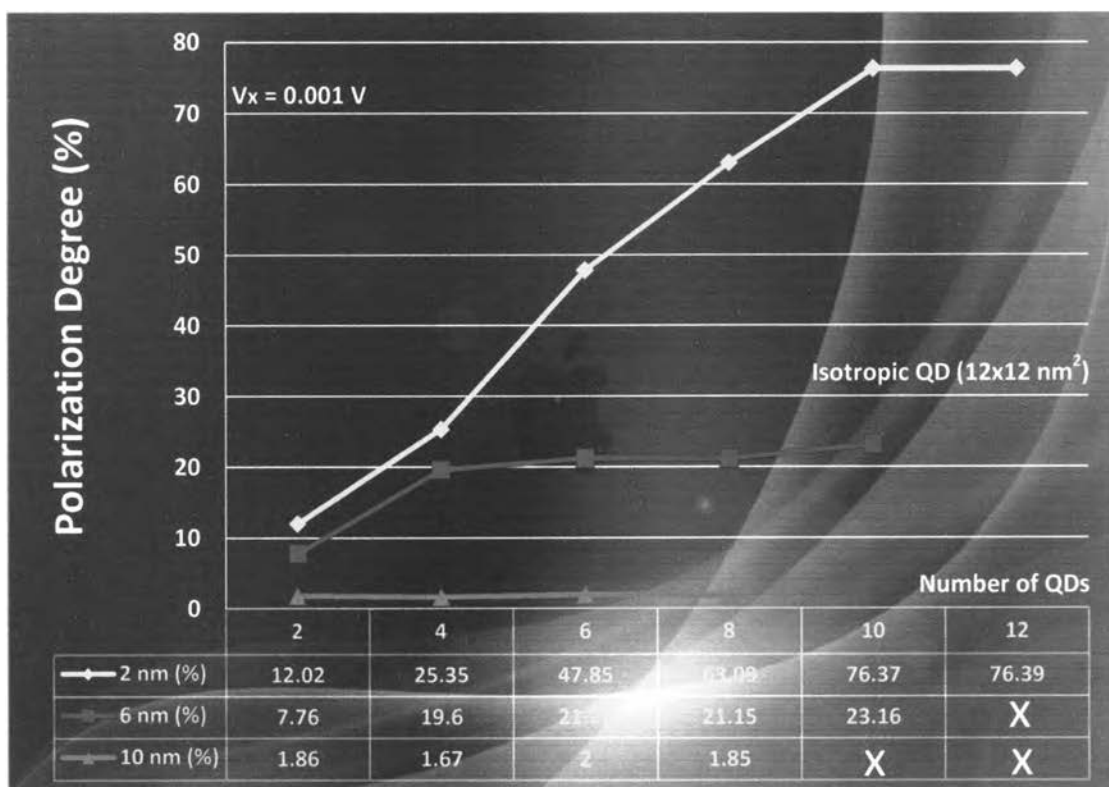
Moreover, the applied voltage causing the polarization degree begins decreasing or " $V_{diminish}$ " (in other words, the first voltage value that greater than  $V_{PD(max)}$  corresponding to this condition) emerges at the lower applied voltage when the number of QDs increases (see **Figure 4.11** and **4.12**). The reason is attributed to the additional carriers (from additional QDs). Although the increase of carriers encourages a high carrier interaction coupling to each other which is larger than

excitons Coulomb interaction, at the same time, when applied voltage  $\geq V_{diminish}$ , closeness of carriers with the same charge (electron-electron and hole-hole interactions) leads to a higher exclusion from each other (due to a stronger repulsive interaction) and also the multiplication of the transition probability in the y direction ( $I_y$  then rapidly goes up). These effects thus contribute to the decrease of polarization degree. It can be interpreted that the smaller of spatial separation between adjacent QDs is required for given a higher polarization degree (both zero and applied electric field cases). Nonetheless, if the system is in the condition of high applied voltage in order to improvement the PD value, it is essential for well-defined interdot spacing to gain the highest degree of polarization as desired.

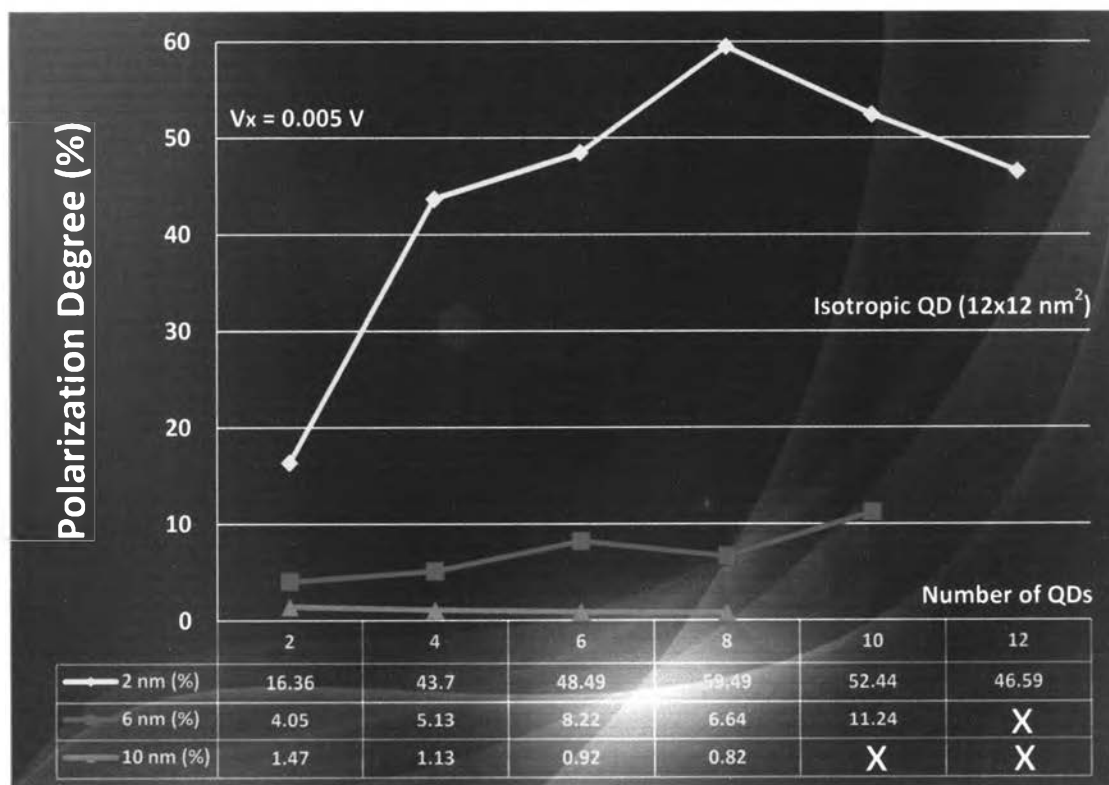
- In the strong applied electric field state ( $> 0.01$  V/nm), the induced electric dipole moment of excitons tends to be saturated due to the confinement of the QDs. The overlarge intensity of electric field completely destructs the excitons Coulomb interaction (no longer an attractive force between carriers) and pushes the carriers to move without a direction reflecting the unstable of the system (now the band structure is out of shape), so it is difficult to indicate which direction of carrier transition happens). This causes  $I_x$  and  $I_y$  are close proximity, inasmuch as, the polarization degree becomes decrease drastically. This biased voltage compelling the PD to begins sharply decreasing is represented as “breakdown voltage” of the system or “  $V_{breakdown}$  ” related to the threshold voltage which produces the polarization degree to converge or lower than the PD value in case of zero field. The decrease of PD by an influence of  $V_{breakdown}$  is obviously seen when the number of QDs increases (the reason is the same as described in the intermediate applied field case).

In case of the optical responses, when the electric-field potential is larger than the exciton binding energy, the field overcomes Coulomb interaction and the optical responses decrease greatly. The existence of applied electric field gains a higher number of excited electrons and holes (resulting in a broadening of absorption spectra) increasing the probability of carriers tunneling out of the quantum well (reduces the excitons lifetimes), leads to an increasing of optical polarization. However, if the electric field is "very strong", it affects the electrons and holes to widely separate from each other which limits their optical responses (since excitons lifetimes return to increases), as well. This emphasizes an important of electric field strength on the polarization degree of QD system.

As previously discussed in **section 4.2.3**, it reflects that elongated QD structure is more gainful than the QD structure whose size extended in both directions. Despite of the fact that more elongated QD structure is unable to be grown, it can be compensated by aligned QD structure for given a high polarization degree. In brief, when designing optical devices using QDs structure, the dot size and electric field strength should be determined by taking both the polarization characteristics and optical response properties into consideration.



(a)



(b)

**Figure 4.13** The polarization degree of  $12 \times 12$  nm<sup>2</sup> QDs vs. the number of QDs at three different interdot spacing of 2, 6, and 10 nm respectively under two constant applied voltages of (a) 0.001 V and (b) 0.005 V.

### 4.3.2 Variation of Polarization Degree with Interdot Spacing under Electric Field

In **section 3.1.4.2.2**, the calculated results reveal a decrease of polarization as increases a distance between dots. The purpose of this topic is to inquire the influence of applied electric field whether it can enhance polarization degree even in a large interdot separation or not. The polarization degree of  $12 \times 12 \text{ nm}^2$  (isotropic shape) QDs versus the number of QDs at three different interdot spacing of 2, 6, and 10 nm respectively under two constant applied voltages, is illustrated in **Figure 4.13**. Note that a cross symbol shown in this figure represents PD value in the condition that couldn't be estimated because it is out of limitation in calculation process (out of memory). At  $V_x = 0.001 \text{ V}$  (**Figure 4.13 (a)**), the tendency in case of interdot spacing of two and six nm show an increasing of PD values while increasing the number of QDs, but decreasing when the separation is 10 nm. Another result at  $V_x = 0.005 \text{ V}$  (**Figure 4.13 (b)**) is inadequate for an analysis to go on due to variance of PD values. We therefore re-calculated by using the same QD size with specification of four different interdot spacing about 2, 6, 10, and 18 nm, respectively under the applied voltage varied from 0 to 0.005 V, as depicted in **Figure 4.14**. Three different cases of QDs: 2, 4, and 6 QDs are selected for comparison.

In binary QDs structure, a demonstration can be considered by separating in four periods of distance between adjacent QDs, and the PD results (**Figure 4.14 (a1)**) are described as follows:

- at 2 nm interdot spacing, it can be observed that all of the PD values for interdot spacing of 2 nm at this electric field's range systematically increase, as shown in **section 4.3.1**.
- at 6 nm interdot spacing, the PD value increases from 4.59% at zero voltage to 7.76% (green rectangular) at applied voltage of 0.001 V (which is PD maximum)

before orderly decreases as  $V_x > 0.001$  V. An overview of all PD values in this case is smaller than that PD values at the same applied voltage in the previous case (2 nm), even in the PD at zero applied field (9.34%).

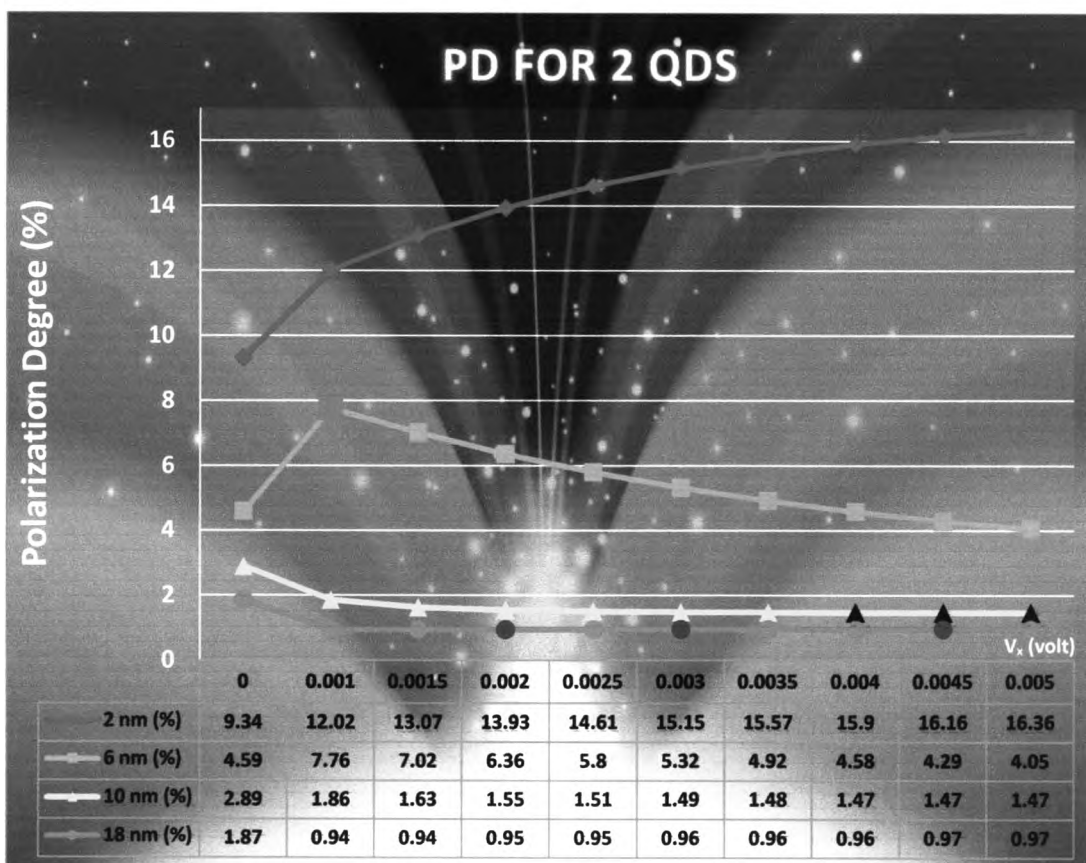
- at 10 nm interdot spacing, a continually reducing of polarization degree arises from an increasing of applied voltage, and become flat when  $V_x$  reaches to 0.004 V (black triangle). The tendency of all PD values is “much” smaller than that in case of both 2 nm and 6 nm.

- at 18 nm interdot spacing, the fluctuation of PD values is observed. The polarization degree is decreasing while applied voltage is in the range of  $0 < V_x < 0.002$  V. After that, the PD value begins increasing at  $V_x = 0.002$  V (PD = 0.95%) called the “turning point” (blue circle), and occurrence of this point can be seen again at  $V_x$  of 0.0035 (PD = 0.96%), and 0.0045 V (PD = 0.97%) respectively. In addition, all PD values is slightly different compared to the PD in case of 10 nm interdot separation.

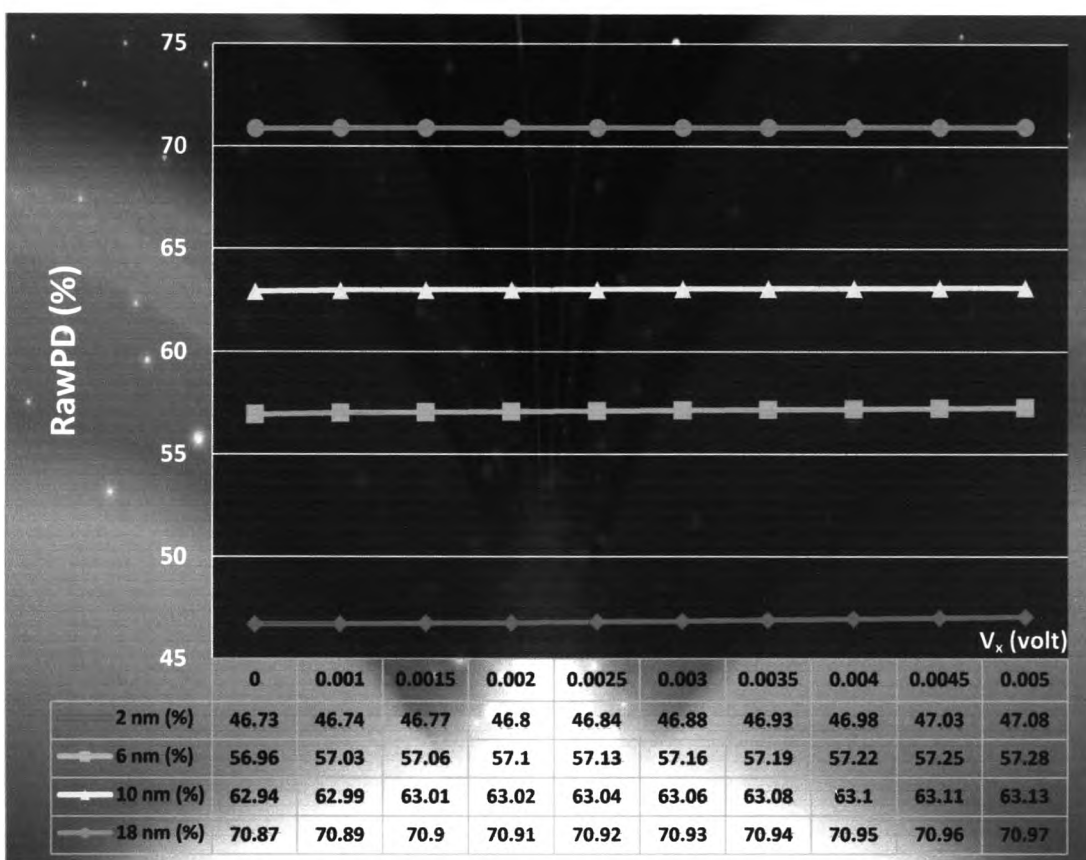
(enlarge on the results mentioned above, some noticeable symbols used in this section refer to connotation as follows. A green symbol is the PD maximum point corresponding to  $V_{PD(\max)}$ , black symbol means a polarization degree that is nearly a constant value even in the increasing of applied field, and blue symbol displays a turning point (here, a turning point stands for a point that PD value returns to increase again))

The explanation of above calculation of PD results and comparison with the case of higher number of QDs (**Figure 4.14 (b1) and (c1)** for 4 and 6 QDs, respectively) are illuminated hereinafter. The results show a polarization degree, which is still inversely proportional with interdot separation even in the assistance of applied electric field (same as the results shown in **section 3.1.4.2.2**). The shift of  $V_{PD(\max)}$  to lower voltage energy when distance between dot increases from 2 to 6

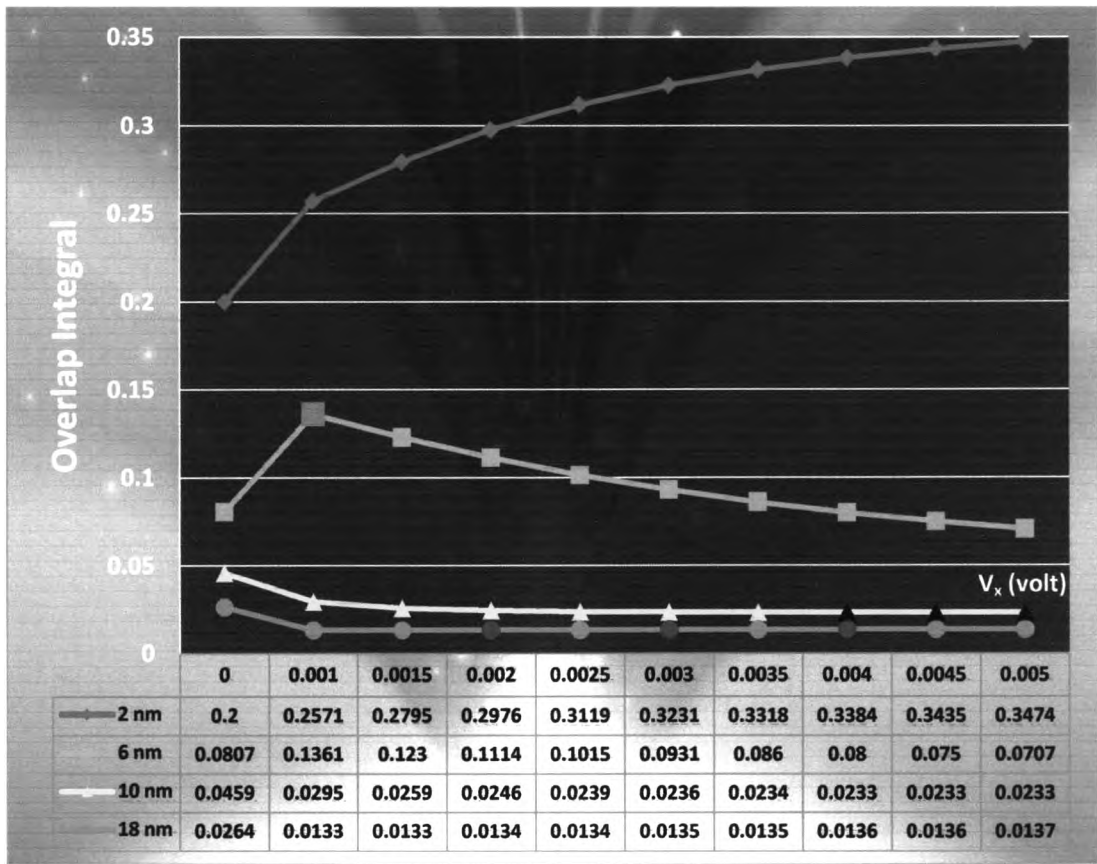




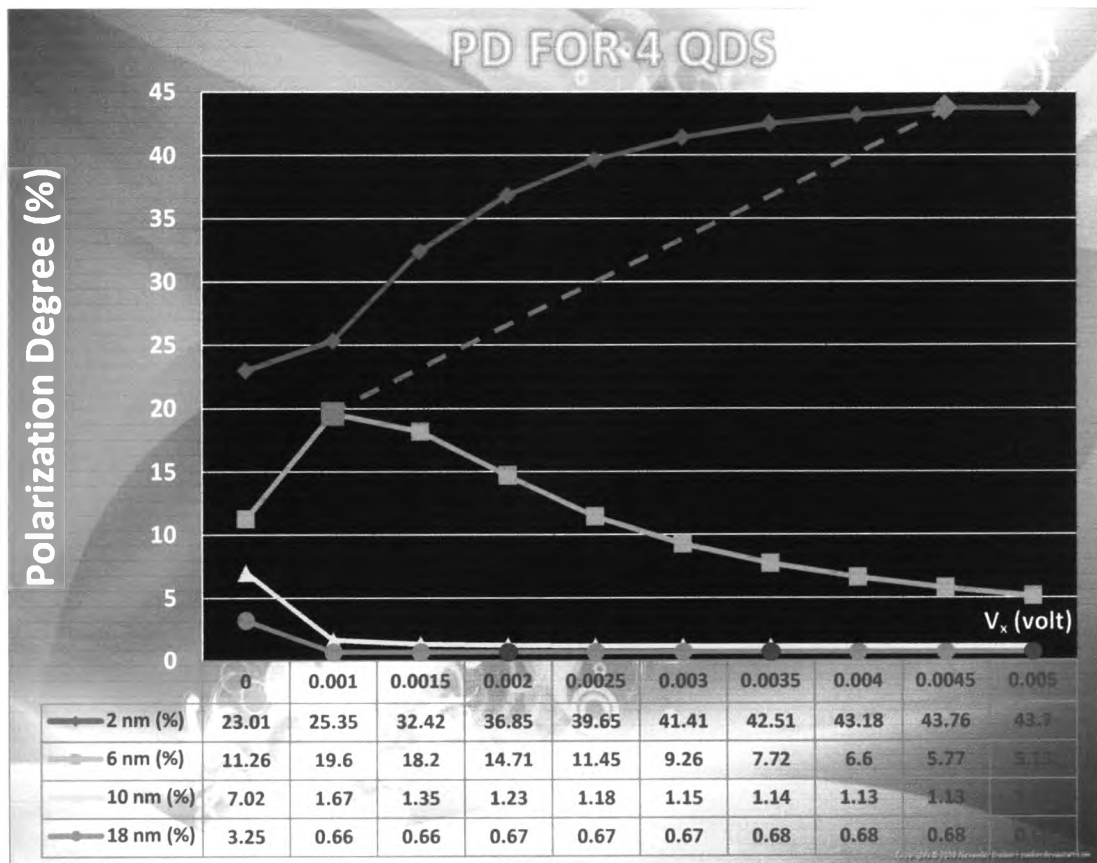
(a1)



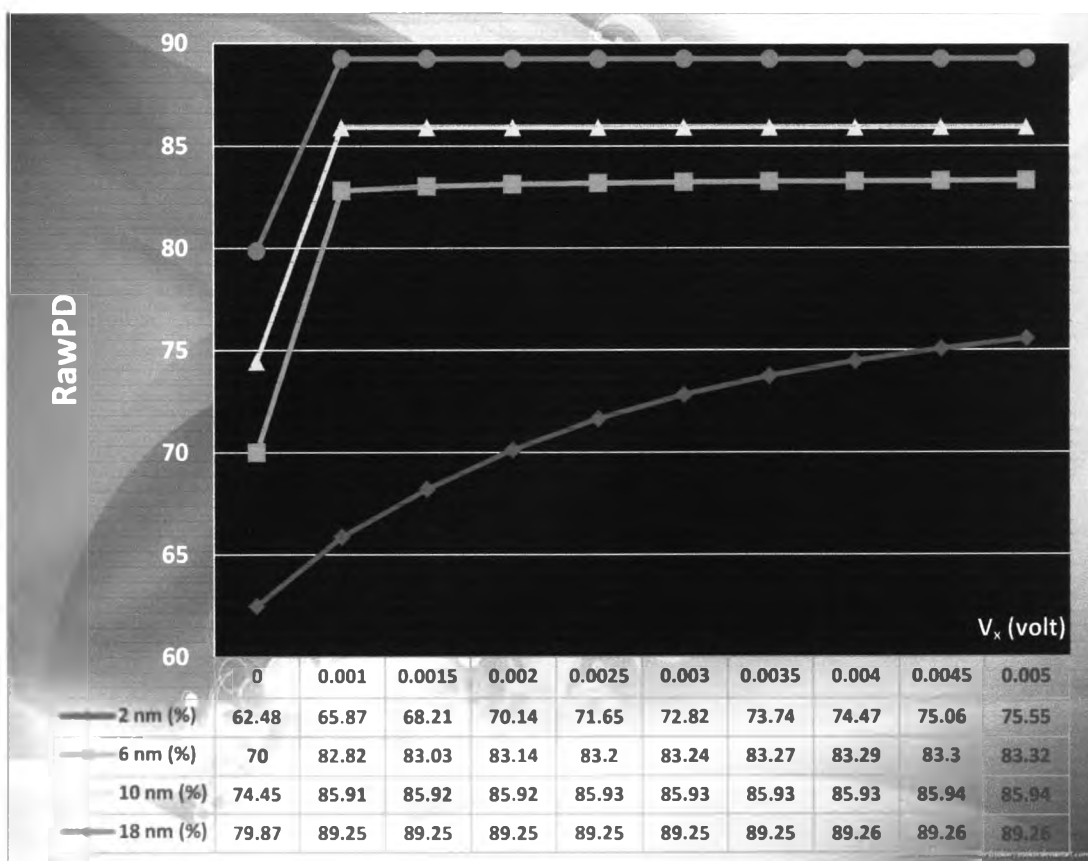
(a2)



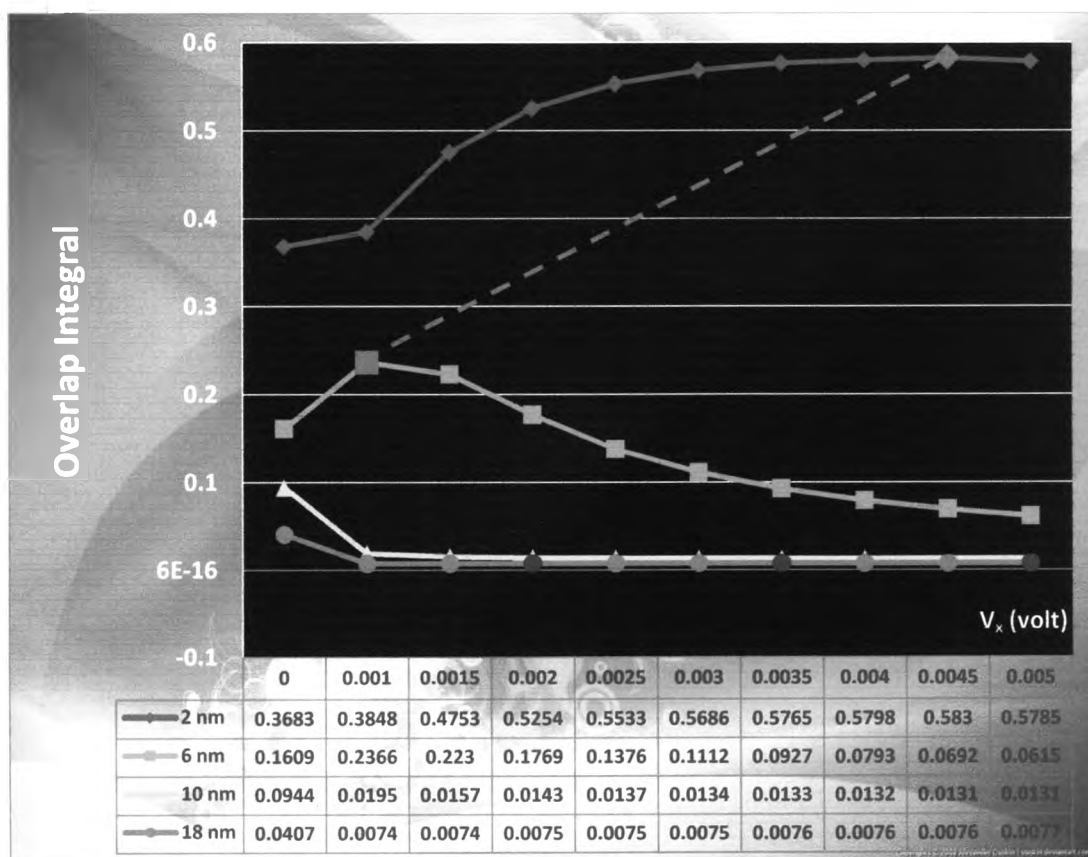
(a3)



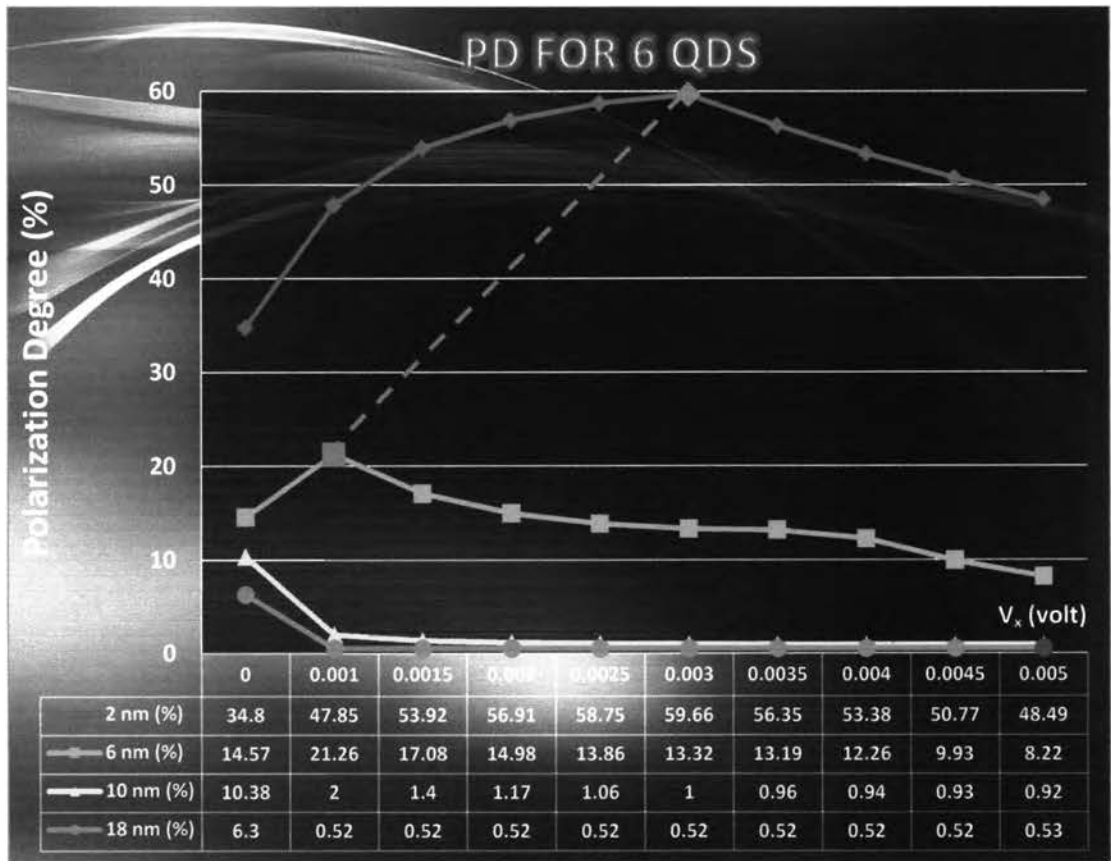
(b1)



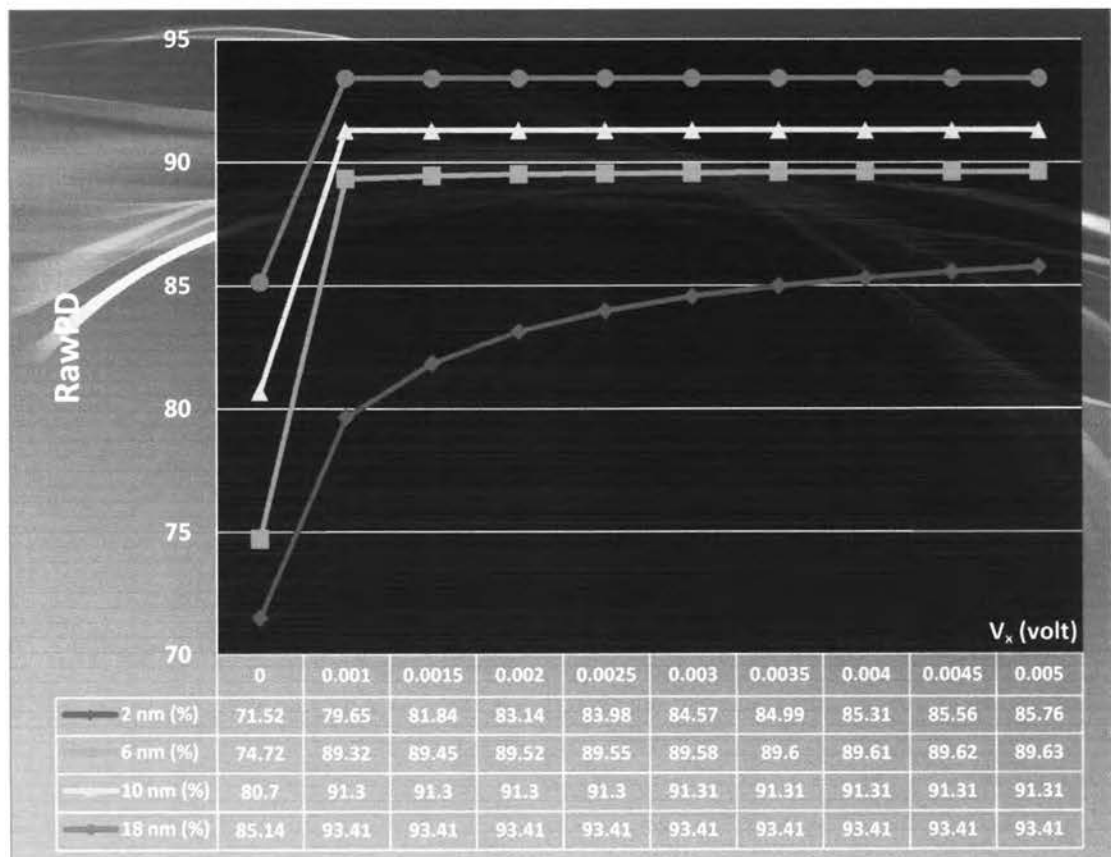
(b2)



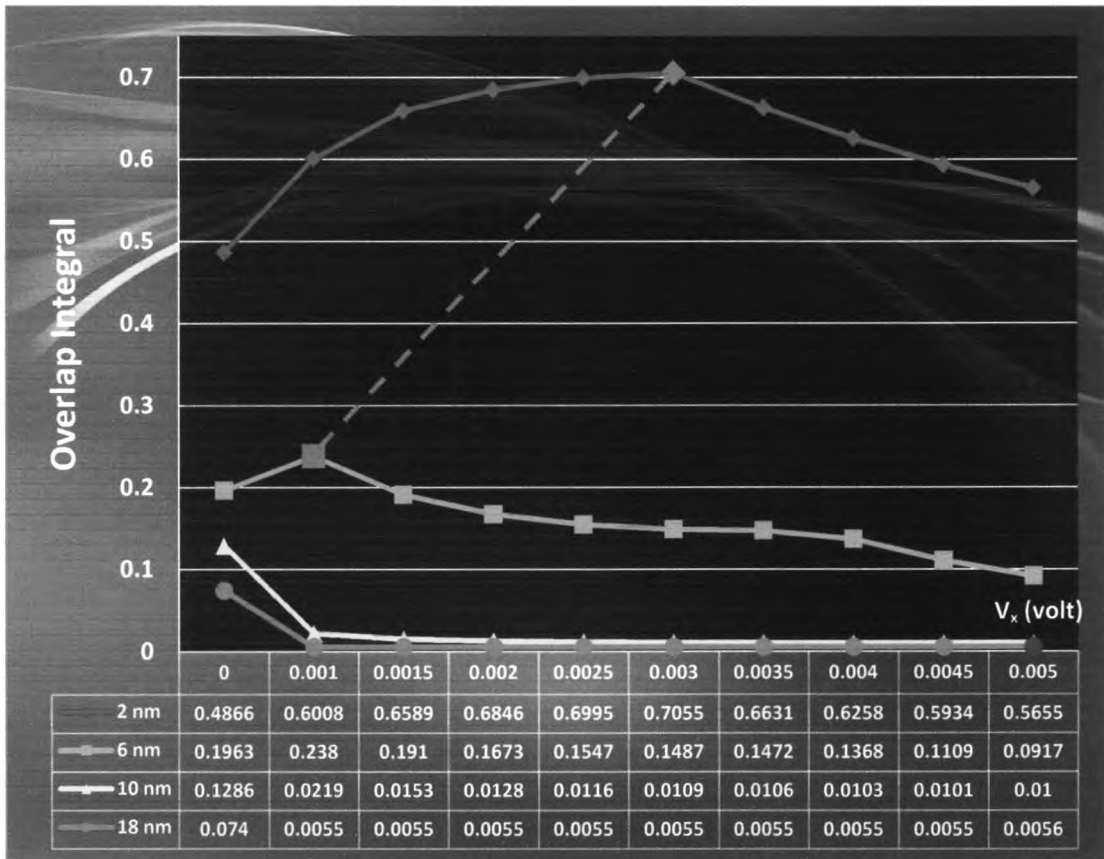
(b3)



(c1)



(c2)



(c3)

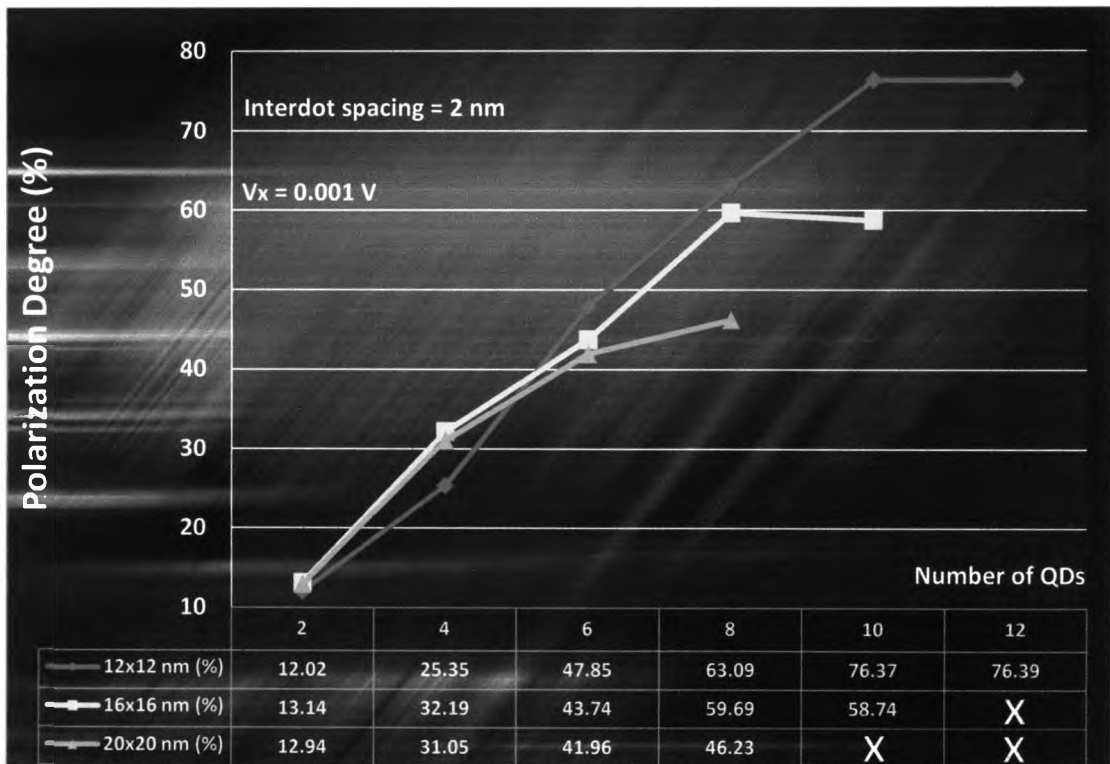
**Figure 4.14** The polarization degree of  $12 \times 12 \text{ nm}^2$  (a1) 2, (a2) 4, and (a3) 6 QDs vs. applied voltage varied from 0 to 0.005 V at four different interdot spacing of 2, 6, 10, and 18 nm respectively. The overlap integral and RawPD corresponding to PD values of (a2 & a3) 2, (b2 & b3) 4, and (c2 & c3) 6 QDs, respectively.

nm is clearly seen (linked by green dashed line) in case both 4 (0.0045 V  $\rightarrow$  0.001 V) and 6 dots (0.003 V  $\rightarrow$  0.001 V), but not emerge in case of binary dots (and also not appears when increases from 6 (10) nm to 10 (18) nm in all QD cases). The tendency of PD values in case of 4 and 6 QDs at interdot spacing both 2 nm and 6 nm remains higher than that the case of 2 QDs. However, at larger interdot spacing (10 and 18 nm) PD values in case both 4 and 6 QDs turns to decrease rapidly than in case of 2 QDs, and more decrease when increase the number of QDs. Moreover, some PD values of binary and four QD cases at interdot spacing of 10 nm show a constant PD when applied voltage is over 0.004 V (for six QDs, PD value slowly decreases), and at

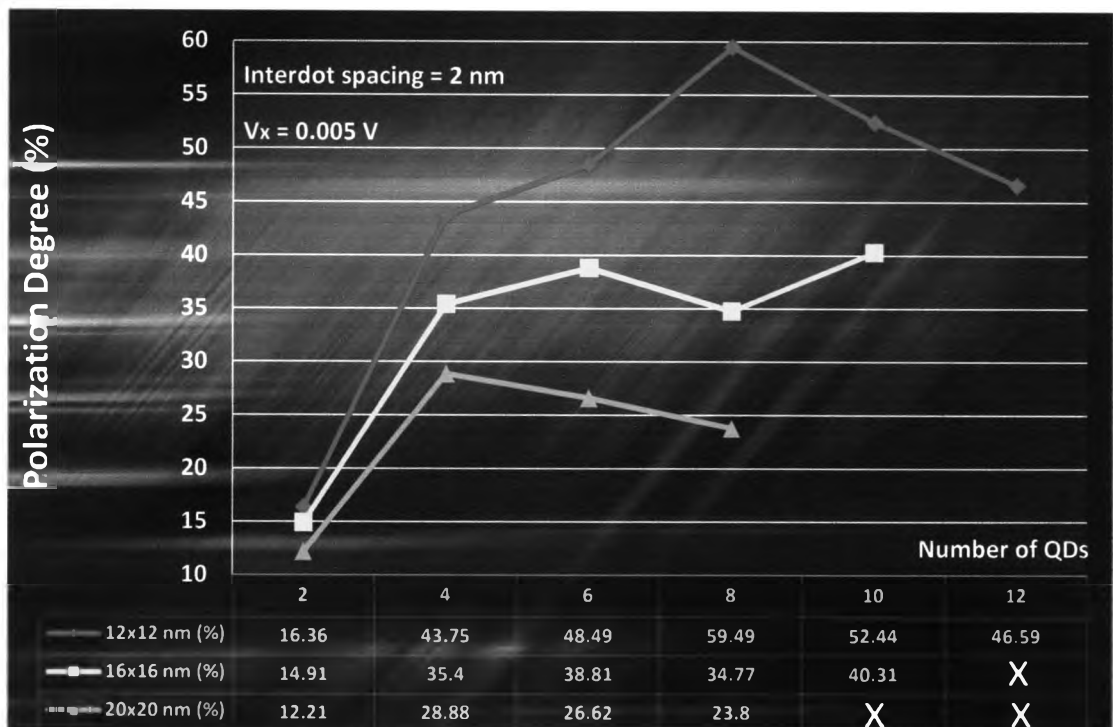
interdot spacing of 18 nm, three turning points are already seen by slightly increase, as well (for six QDs, PD value is constant at all voltages). PD results tell us that PD in all number of QDs produces the highest value when interdot spacing is 2nm.

In the relation of polarization degree with RawPD and overlap integral, RawPD of binary dots (**Figure 4.14 (a3)**) gradually increases when increases an applied voltage. This situation is similar to 4 and 6 QD cases (**Figure 4.14 (b3)** and **(c3)**, respectively), but slightly different as  $V_x$  increases from 0 to 0.001 V (PD value increases rapidly) since the higher number of QDs displays a better response with an increase in range of low applied electric field. For overlap integral, the results show that their tendency are the same as variation of polarization degree for all QD cases. We stress again as mentioned previously in **section 4.3.1**, electric field insignificantly affect on changing of RawPD meanwhile an outstanding variation occurs at the overlap integral values. In fact, a crucial factor for determining the total polarization degree of QD system is the “overlap integral” term representing as “multiplier”, which leads to numerous fluctuation of PD value.

According to the calculated results, the large separation between QDs, the greatly decrease of overlap integral then manifests (this cause an interdot spacing far enough so that interaction of carriers is very low). Even if electric field is applied in order to adjust PD value become higher, nevertheless, it is still inadequate in the case of large dot separation which in “dilemma” for improvement the PD value. On the contrary, the effect of electric field encourages a drastically decrease of overlapping between carrier wavefunctions, resulting in a small polarization degree, as well. Hence, it can shortly make a conclusion that the condition of suitably distance between QDs must be satisfied to give a strong carriers interaction for the high polarization of QD system.



(a)



(b)

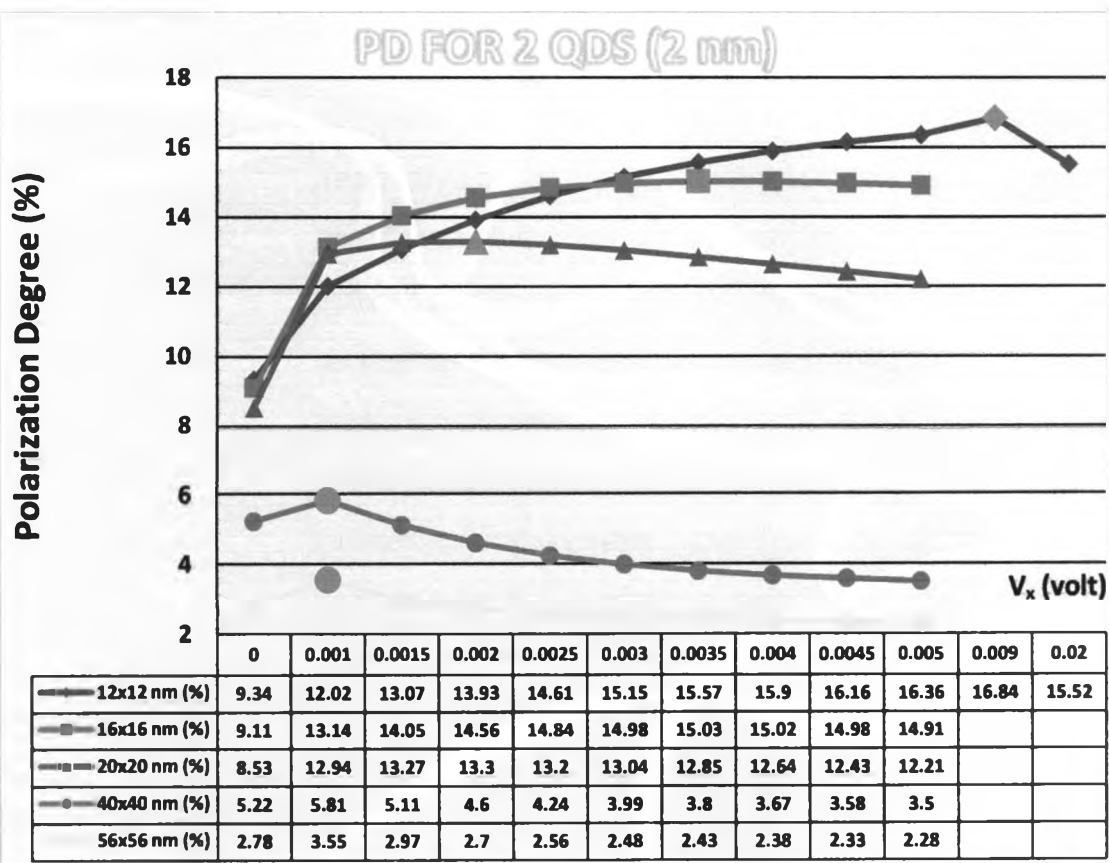
**Figure 4.15** The polarization degree vs. the number of QDs at three different QD sizes of 12x12, 16x16, and 20x20 nm<sup>2</sup> respectively under two constant applied voltages of (a) 0.001 V and (b) 0.005 V. An interdot spacing was fixed to 2 nm.

### 4.3.3 Comparison of Polarization Degree for Various QD Sizes in Presence of Electric Field

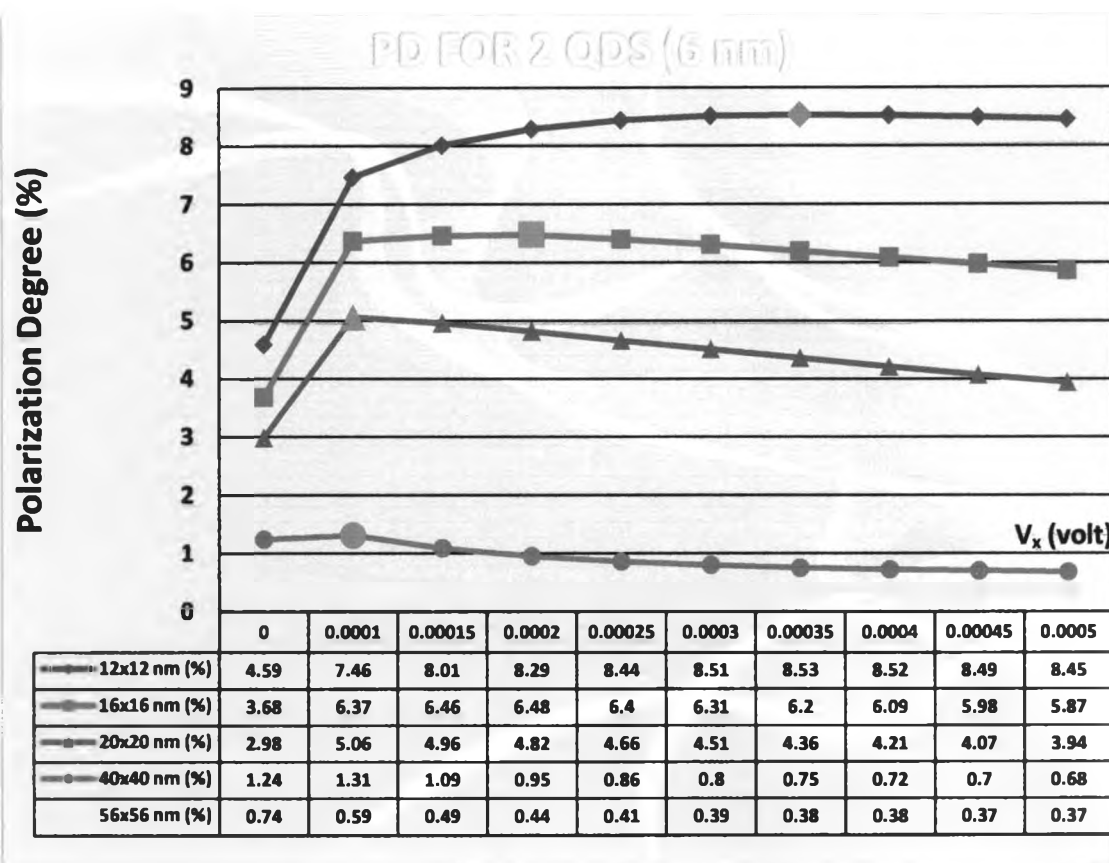
Remember in **section 4.2** that the results give details about “single QD” in view of higher PD value when QD size is larger under an “applied electric field”, at the same time, the calculation in **section 3.1.4.2.2** shows PD increasing as decreasing the QD size in “aligned QD” structure (high probability in overlapping of carrier wavefunctions) at “zero field”. Between two contradicting results, how does extending QD size in all direction affect on the polarization degree of aligned QDs in circumstance of applied field? This last section will thus give an answer for clarification. **Figure 4.15** shows the variation of polarization degree versus number of QDs (interdot spacing was fixed to 2 nm) at three different isotropic QD sizes of 12x12, 16x16, and 20x20 nm<sup>2</sup>, respectively, under two constant applied voltages. The polarization degree of all cases goes up when the number of QDs increases at applied voltage equal to 0.01 V (**Figure 4.15 (a)**), showing a good tendency for higher polarization. However, at  $V_x = 0.005$  V (**Figure 4.15 (b)**), there is no sufficient information to predict the change in PD.

This problem can be resolved by measuring the polarization degree again, but choosing three different numbers of QDs, that is, 2, 4, and 6 QDs, and extending to five different QD sizes: 12x12, 16x16, 20x20, 40x40, and 56x56 nm<sup>2</sup>, respectively. For 4 QDs, the QD sizes of 40x40, and 56x56 nm<sup>2</sup> were replaced by 28x28, and 40x40 nm<sup>2</sup>, respectively, due to limitation in calculation process. In 6-QD case, only three different QD sizes were chosen: 12x12, 20x20, and 24x24 nm<sup>2</sup> (same reason in the case of 4 QDs). We also separated into two different interdot spacing: 2 nm corresponding to applied voltage varied from 0 to 0.005 V, and 6 nm corresponding to applied voltage varied from 0 to 0.0005 V (the voltage in the latter case is ten times lower than the previous one to clearly observe the variation of PD). Note that the RawPD and overlap integral results are not shown because their tendency is the same as described in **section 4.3.2**.

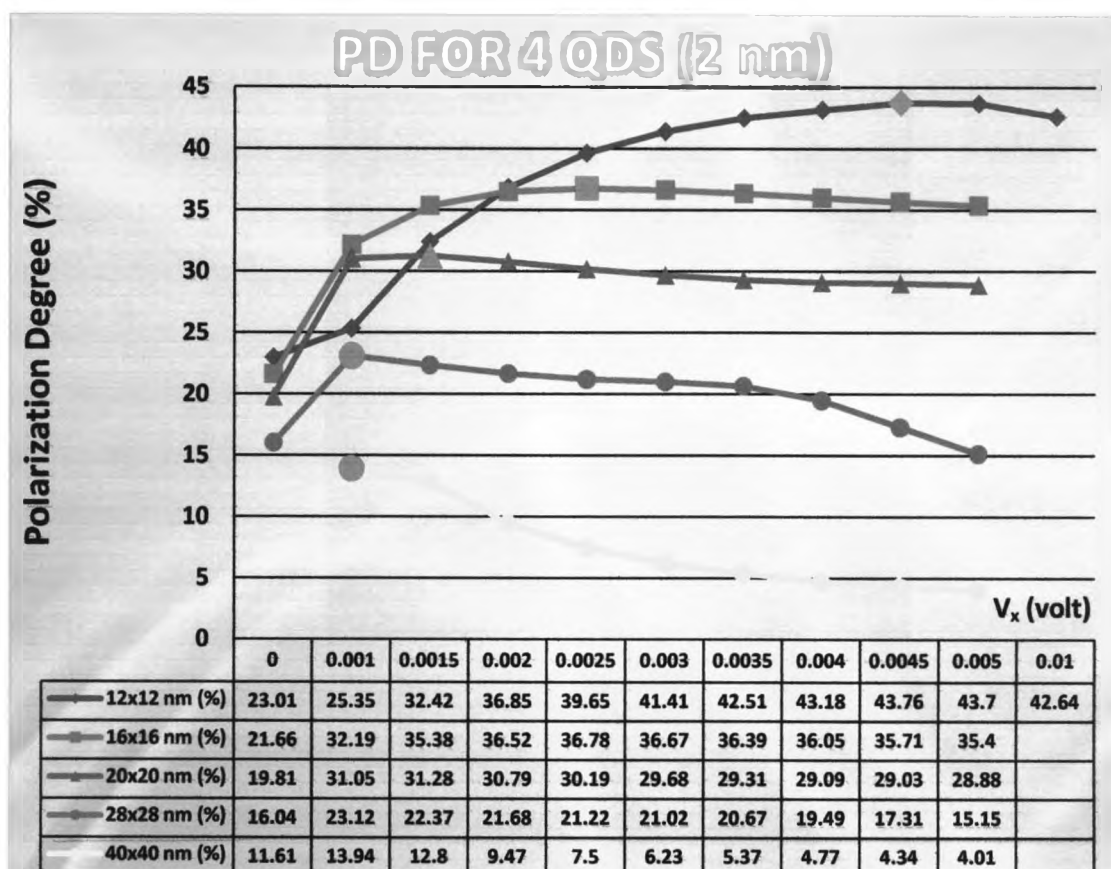




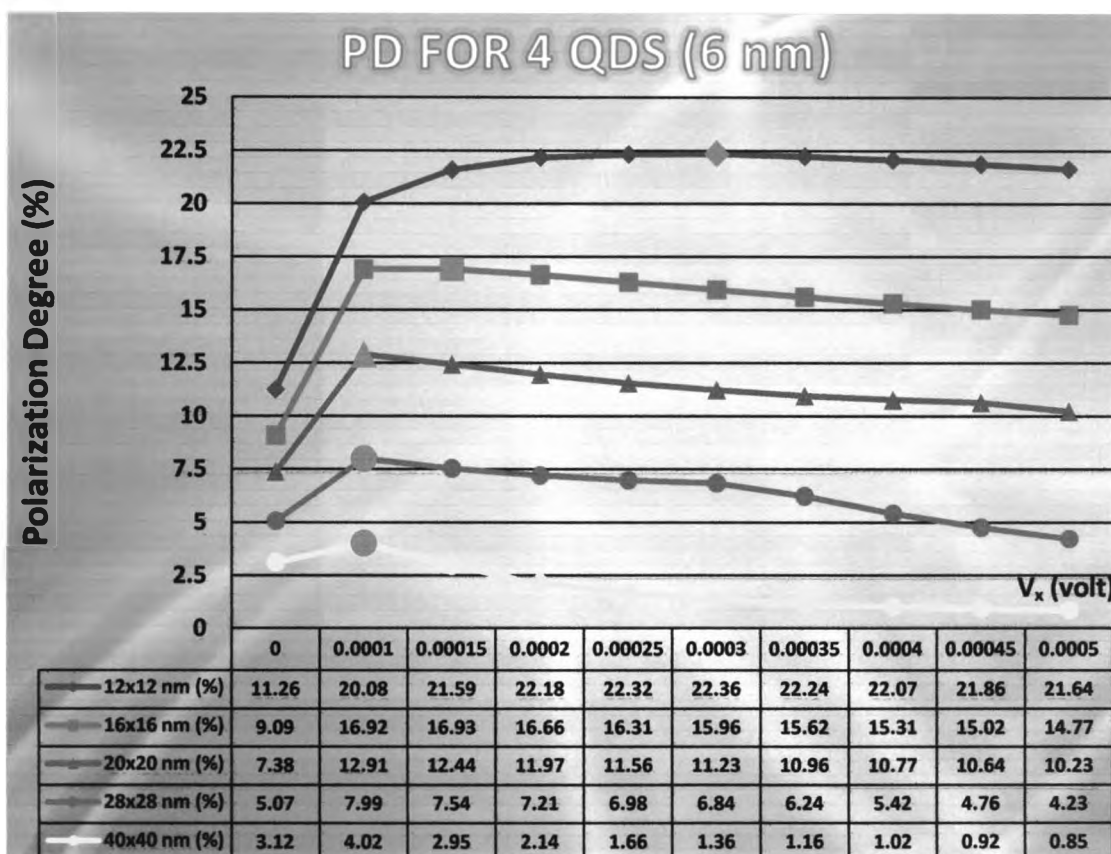
(a1)



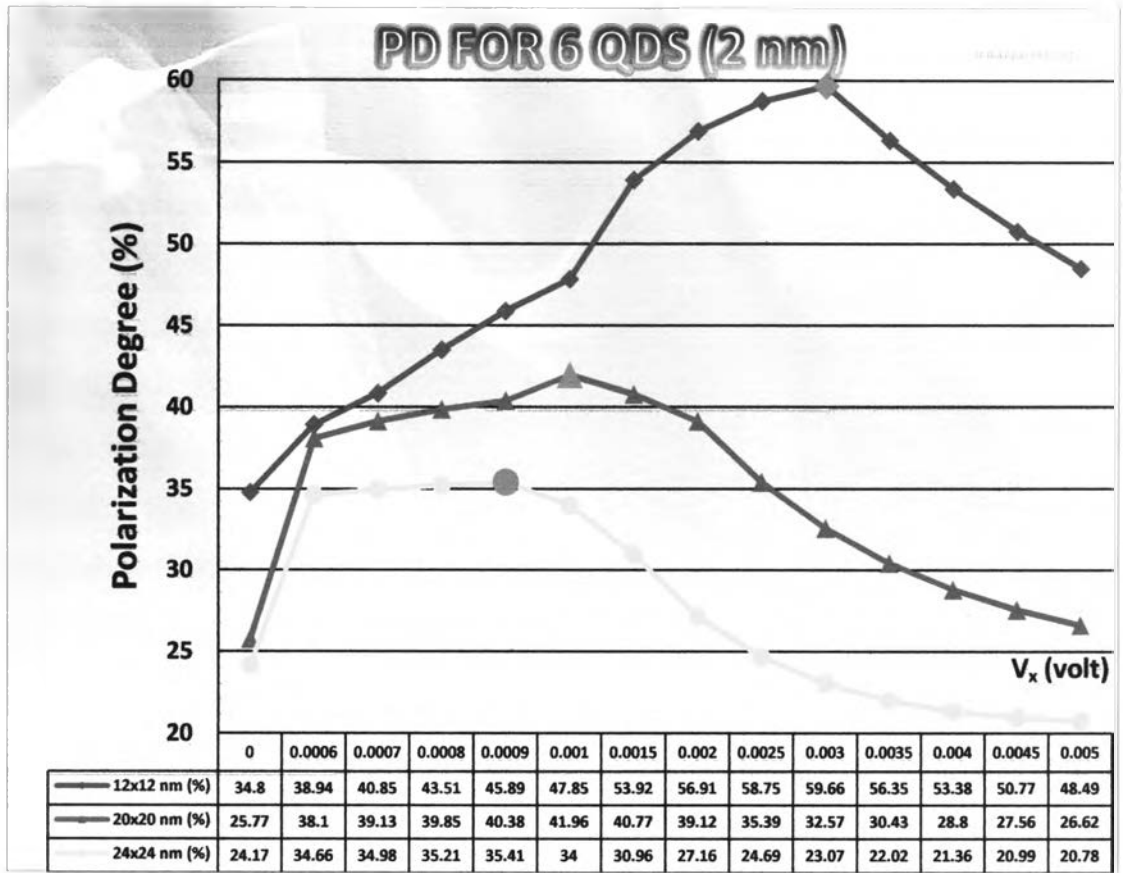
(a2)



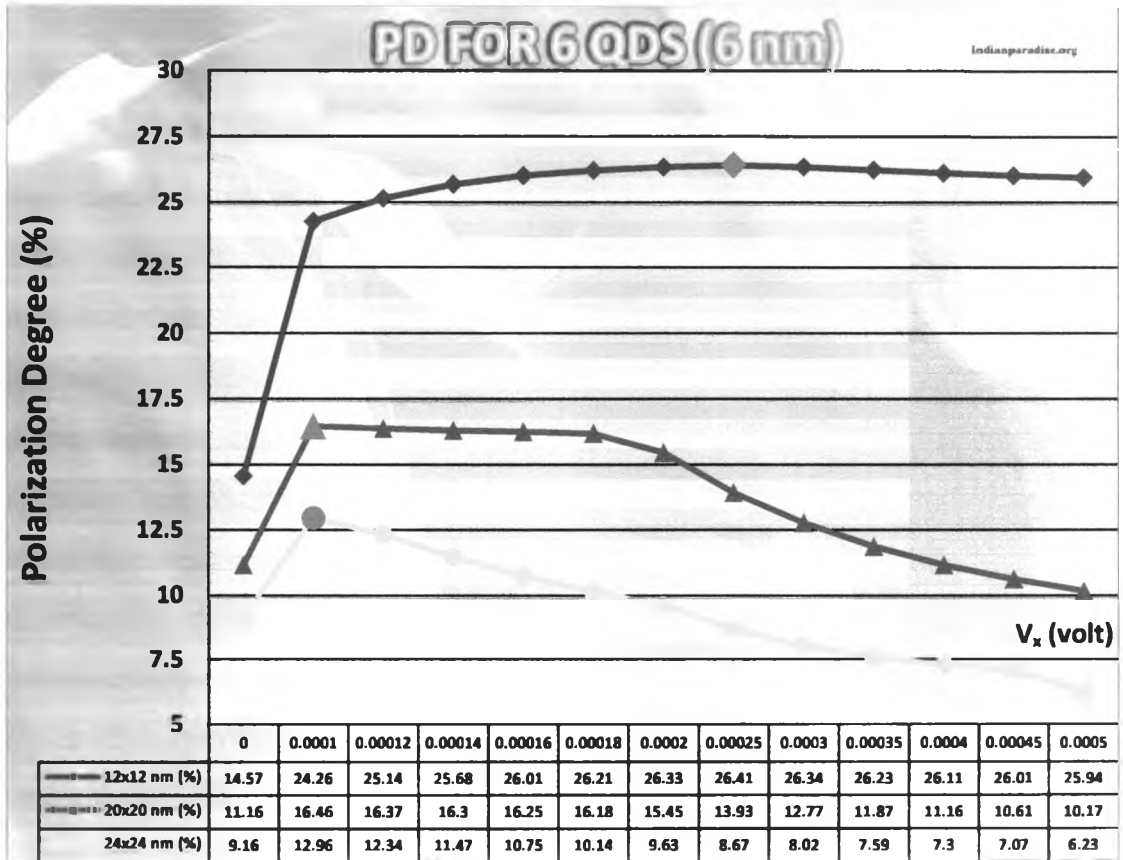
(b1)



(b2)

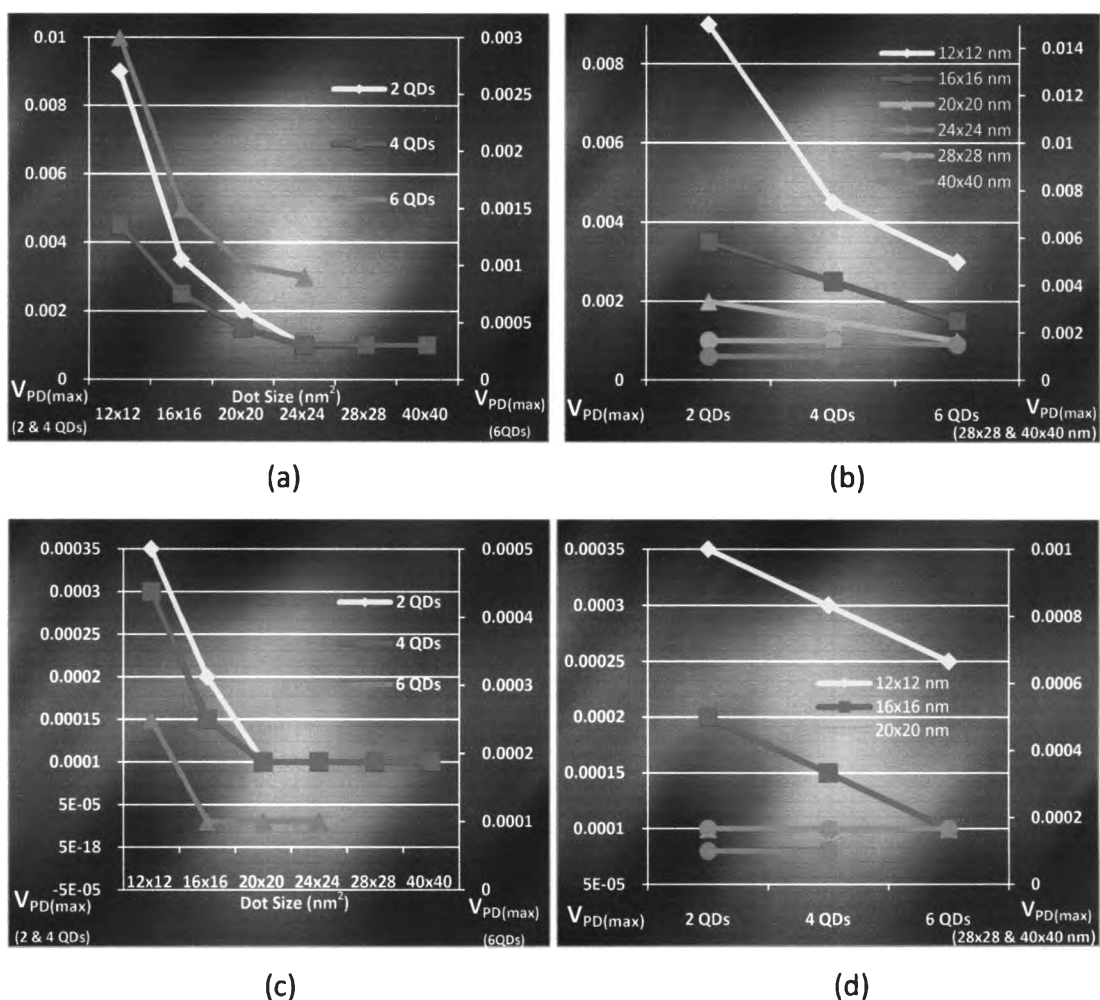


(c1)



(c2)

**Figure 4.16** The polarization degree vs. applied voltage of five different QD sizes: 12x12, 16x16, 20x20, 40x40, and 56x56 nm<sup>2</sup>, respectively (for 4-QD case, the QD sizes of 40x40, and 56x56 nm<sup>2</sup> were replaced by 28x28, and 40x40 nm<sup>2</sup>, respectively, and for 6-QD case, there were three different QD sizes: 12x12, 20x20, and 24x24 nm<sup>2</sup>) at two conditions of interdot spacing: 2 nm corresponding to applied voltage varied from 0 to 0.005 V of 2 (a1), 4 (b1), and 6 (c1) QDs, respectively, and 6 nm corresponding to applied voltage varied from 0 to 0.0005 V of 2 (a2), 4 (b2), and 6 (c2) QDs, respectively.



**Figure 4.17** The  $V_{PD(max)}$  vs. QD sizes of 12x12, 16x16, 20x20, 24x24, 28x28, and 40x40 nm<sup>2</sup> with three different numbers of QDs: 2, 4, and 6 QDs at interdot spacing of (a) 2 nm, and (c) 6 nm, respectively. The  $V_{PD(max)}$  vs. number of QDs (2, 4, and 6 QDs) with 6 QD sizes mentioned above at interdot spacing of (b) 2 nm, and (d) 6 nm, respectively.

**Table 4.3:** The  $V_{PD(max)}$  vs. QD sizes & number of QDs at interdot spacing of 2 and 6 nm.

$V_{PD(max)}$ (V)	2 nm			6 nm		
	2 QDs	4 QDs	6 QDs	2 QDs	4 QDs	6 QDs
12x12 nm <sup>2</sup>	0.009	0.0045	0.003	0.00035	0.0003	0.00025
16x16 nm <sup>2</sup>	0.0035	0.0025	0.0015	0.0002	0.00015	0.0001
20x20 nm <sup>2</sup>	0.002	0.0015	0.001	0.0001	0.0001	0.0001
24x24 nm <sup>2</sup>	0.001	0.001	0.0009	0.0001	0.0001	0.0001
28x28 nm <sup>2</sup>	0.001	0.001	X	0.0001	0.0001	X
40x40 nm <sup>2</sup>	0.001	0.001	X	0.0001	0.0001	X

Figure 4.16 illustrates polarization degree with the condition corresponding to the calculation reported above. The associated discussion of these results is based on classification by the distance between QDs. In each case, the number of QDs is also taken into account, and will be explained hereinafter.

- At the distance between QDs of 2 nm, several phenomena happen as follows:
  - The polarization degree for all QD sizes and all numbers of QDs shows the same tendency, that is, polarization degree increases when an applied voltage is increased and reaches to the maximum value, corresponding to  $V_{PD(max)}$  (displayed as a green symbol, and same meaning as described in section 4.3.2), then starts to decrease as  $V_x \geq V_{diminish}$ .
  - For 2 and 4 QDs, at low applied voltage ( $V_x < 0.0015$  V) PD values in case of 16x16 and 20x20 nm<sup>2</sup> QDs are higher than that of 12x12 nm<sup>2</sup> QDs. On the contrary, its PD values turn out to be higher than the two previous cases when  $V_x \geq 0.003$  V (or better response to a larger field than those). In larger QDs (40x40 & 56x56 nm<sup>2</sup> for 2 QDs, and 28x28 & 40x40 nm<sup>2</sup> for 4 QDs, respectively), it was found that the PD

values decrease as size of QDs increase. This effect is clearly seen in case of 6 QDs and even more obvious when a strong field is applied.

- Consider  $V_{PD(max)}$  with respect to the QD sizes (**Figure 4.17 (a)**, which we can also see the variation of  $V_{PD(max)}$  in **Table 4.3** by inspecting along a column of three number of QD cases). The shift of  $V_{PD(max)}$  to a lower voltage when the QD size is increased may be observed (and  $\Delta V_{PD(max)}$  in each increase of QD size decreases (or slower shifting) too). However, this effect fades away as QD size is much larger (from 24x24 to 40x40 nm<sup>2</sup> in case of 2 and 4 QDs , showing a constant  $V_{PD(max)}$  of 0.001 V).

- On the other hand, the shift of  $V_{PD(max)}$  with respect to the number of QDs (**Figure 4.17 (b)**, which we can also see for the variation of  $V_{PD(max)}$  in **Table 4.3** by inspecting along a row of six various QD sizes) also decreases to a lower voltage as the number of QDs increases, that is,  $V_{PD(max)}$  for all QD sizes of lower number of QDs is less than  $V_{PD(max)}$  of higher number of QDs, for the same QD sizes (and  $\Delta V_{PD(max)}$  in each increment of number of QDs decreases, too). Besides, when QD size is much larger (from 24x24 to 40x40 nm<sup>2</sup> in 2 and 4 QD cases),  $V_{PD(max)}$  is a constant value of 0.001 V, and slightly changes to 0.0009 V in case of 6 QDs for QD size of 24x24 nm<sup>2</sup>. We therefore conclude that the shift of  $V_{PD(max)}$  at a large QD size is low and becomes very low when the number of QDs increases.

- For the distance between QDs of 6 nm, slightly different situations will be described as below:

- The tendency of polarization degree is mostly the same as the PD results in case of 2 nm interdot spacing (all PD values in this case is smaller than the PD values of 2 nm case for the same magnitude of applied voltage. This is due to the interdot spacing effect, as previously described in **section 4.3.2**), except in 28x28 and 40x40 nm<sup>2</sup> of 2

QDs that PD is always decreasing for an increasing of applied voltage (and more decreasing when the dot size increases).

- The results about shift of  $V_{PD(max)}$  with respect to both QD size and number of QDs are also the same as 2 nm interdot spacing, but slower shifting (and also very small  $V_{PD(max)}$ , seen from the many times of  $V_{PD(max)}$  value of 0.0001 V). Repeating again, it is necessary to reduce a range of applied voltage for 6 nm interdot spacing in order to more clearly observe the  $V_{PD(max)}$  shift.

Now, all results discussed above give us the clarification of the effect of QD size on the polarization degree under applied electric field. Despite the fact that size enlargement of single QD at increasing an applied field produces a higher polarization anisotropy, this turns into “deleterious effect”, representing an obstacle on the increase of polarization in case of aligned QDs because extension of QD size in this condition causes the decrease of PD value. As mentioned earlier in **section 4.3.2** for aligned QD structure, the overlap integral is an important factor to determine the total polarization degree of QD system. Large dimension of the QDs causes the overlap between carrier wavefunctions to decrease due to more localization. The higher number of QDs with large size, the smaller coupling between QDs therefore occurs (already described in **section 3.1.4.2.2**).

A larger QD allows for an increasing of polarization degree in case of lower magnitude of electric field. Because a large QD affects the response of applied field to a low magnitude of  $V_{PD(max)}$ , which is normally less than 2 - 4 times (or more) compared with the smaller QD case. A small  $V_{PD(max)}$  (low applied field) in this case does not produce a high maximum PD value as in case of higher number of QDs (**Figure 4.11** and **Figure 4.12**), but creates a maximum PD value which is only slightly different from the case of no applied field instead. Hence, enlargement of QD size not only insignificantly increases a polarization anisotropy with a small applied field,

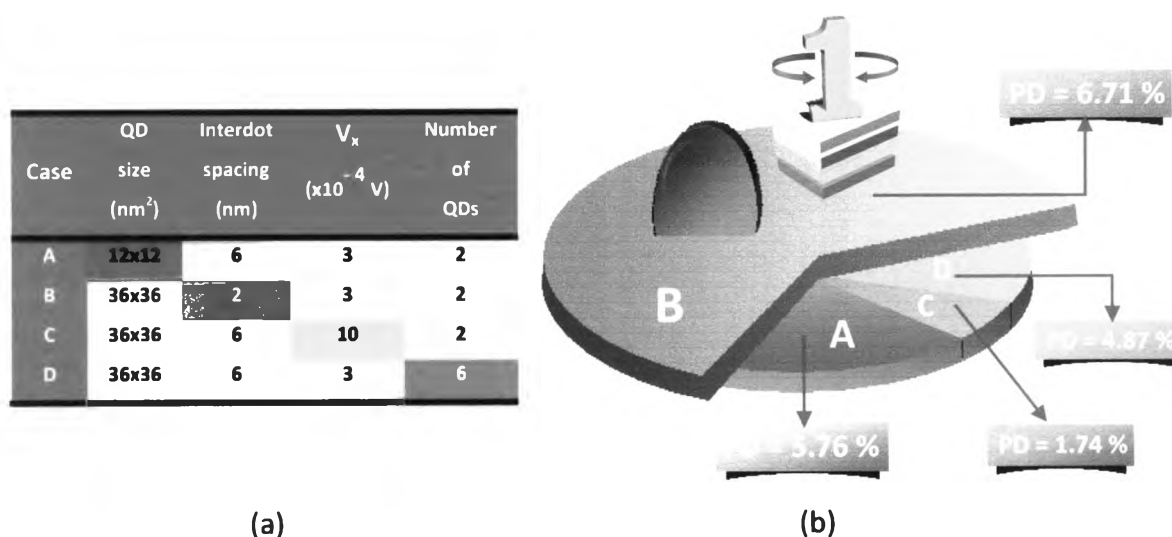
but also rapidly decreases a polarization anisotropy when strong electric field is applied (unlike in case of single larger QD, for which the PD is higher when using a strong field), resulting in no increase of polarization anymore.

Recall from **section 4.2.3** and **4.3.1** that a large QD under “strong” electric field produces a decrease of an optical response even though the polarization degree is higher. The results from this topic also encourage us to avoid the growth of overlarge size of QDs in case of aligned QD structure with an applied field, which limits an increase of both the polarization degree and the optical response as well. In general, fabrication of QDs mostly forms an anisotropic QD shape (and also have some optical anisotropic properties) because of strain between the hetero-interface and the anisotropy in the semiconductor substrate [197], and we expect to get a higher PD than isotropic structures (for which the polarization anisotropy does not occur except in assistance of strong electric field, and even that, low degrees of polarization result).

Nevertheless, the calculated results (not shown here) give smaller PD values for aligned QDs with anisotropic shape than those of isotropic shape under applied field. It may be imagined that an anisotropic shape, even in elongated structures, exhibits polarization anisotropy when they are isolated from others, but in case of aligned QDs their structures have strong confinement which prohibits an overlapping between carrier wavefunctions too (same reason as described in case of large QD). Furthermore, the non-uniformity of QDs is still a problem reported on many research due to complication in fabrication process [197, 231]. This directly affects interdot spacing, overlapping of carrier wavefunctions, and consequently on polarization degree of QD system. Strictly speaking, the aligned QDs with an isotropic shape is satisfactory for giving a strong polarization under applied electric field.



Before the end of this section, we know from all calculated results described in this work that there are several parameters that have been proposed and investigated for high polarization degrees. Among these, which one is the most promising parameter for giving the highest polarization degree of aligned QD system? We therefore investigate by selecting four different cases related to four important parameters: QD size, spacing between QDs, number of QDs, and electric field strength of aligned QDs for comparison. For each QD case, one of those parameters (number of QDs or magnitude of applied voltage) was increased up to three times compared with its original value and the other parameters were fixed (for interdot spacing or QD size, they were decreased down to three times compared with its original value) for producing a higher PD value, corresponding to the conditions as shown in **Figure 4.18 (a)**. Note that an applied voltage in case C stands for  $V_{PD(max)}$  in order to give a maximum PD value for this case. Surprisingly, a parameter that contributes to the highest polarization degree is the “**separation between QDs**” of aligned QD system.



**Figure 4.18** (a) Comparison of polarization degree (in form of table) of 4 different cases: QD size, spacing between QDs, number of QDs, and electric field strength of aligned QDs (the highlighted rectangular represents an adjusted parameter in each case). (b) The pie chart of PD results.

This is an evidence that the interdot spacing most influences the polarization anisotropy since it gives rise to very large variation of the overlap integral and the PD value. Consequently, an amount of interaction between QDs plays as a “prominent factor” evaluating an existence of polarization, as well. In the contrary, an applied electric field just plays as a “supplementary” to fine-tune an interdot coupling for giving a larger the polarization degree.

Although the effect of interdot spacing is outstanding than the others, we should not “disregard” those other effects. If we focus only on the improvement of separation between dots by neglecting them (especially the effect of increase the number of QDs which also directly affects on the increase of polarization degree), this causes a limitation for high degree of polarization anisotropy and sometimes suppresses the performance when operating as an electronic device (because other effects that we overlook them may lead to decreasing of efficiency, so we need to modify and compensate them). Therefore, it is necessary to optimize these factors for approaching to a perfect polarization degree, and provide a high potential electro-optical devices.

## 4.4 OVERALL DISCUSSION

In summary, the main issues from all calculated results in this chapter are repeated and presented as below:

1. First, the two-dimensional InAs linearly aligned QD structure in presence of two-dimensional applied electric field system (controllable via applied voltage) was simulated for all cases of calculations.

2. In the case of electric field's direction, it was shown that the applied electric field in the direction parallel to the direction of elongation or alignment of QDs strongly affect the variation of polarization degree of aligned QDs than the applied electric field in the perpendicular direction. This was due to the fact that  $E_x$  caused more difference between the transition probability in both directions by promoting the carriers (from band tilting) to tunnel across potential barrier (between adjacent QDs) along the directions of QDs alignment so that the polarization degree increases. From these results,  $E_x$  was selected for the rest of the calculations.

3. In consideration of single QD structure, we found that for an isotropic QD (which originally exhibited almost no polarization anisotropy), the assistance of applied electric field acted on the electron and hole wavefunctions (related to electron and hole carriers that initially were localized in QD) to separate from each other, and overlapping between them was decreased. The electron wavefunction distributed farther away from the center than the hole wavefunction (because of larger kinetic energy), but the hole wavefunction was more distorted than the electron wavefunction (which is more easily observed from its spatial asymmetry), so changing of the hole wavefunction was important to determine optical polarization properties. Nevertheless, the shape of carrier wavefunctions remained spatially symmetric since they were oppositely attracted by Coulomb interaction. An applied field also allowed an incoming photon with energy less than the bandgap energy to

be absorbed, therefore an amount of carriers tunneling out of the well increased (carrier lifetimes decrease), resulting in the manifestation of polarization anisotropy.

Interesting results could be seen from the elongated QD structure: there is strong lateral size dependence when there is an applied electric field, which is related to the QCSE, which produces the carrier movement to be confined in the direction of QD alignment (the major contribution is due to the stronger lateral confinement of the hole). This leads to the transition probability in that direction greater than in the perpendicular direction, which gives rise to the large polarization degree, and to become larger with increased applied field or lateral size of QD.

Comparison about QD aspect ratio, in case of the same aspect ratio, larger QD gives a higher polarization because of the large asymmetry of the wavefunctions (from lower confinement). For the larger QDs in a strong electric field, the hole wavefunction was largely localized, and large optical anisotropy thus occurs. Besides, at the high aspect ratio (or large lateral size), polarization degree also increases (the more elongated structure, the larger electric field-induced anisotropy was obtained).

4. The polarization degree of aligned QDs was investigated (extended from the case of single QD). Calculation results amazingly show that variation of polarization degree strongly comes from electric field strength, which may be described as follows:

- In the low magnitude of applied electric field, QCSE induces more interdot coupling and overlapping of carrier wavefunctions thus increases, leading to the increase of polarization degree from the initial case of no applied field. The PD values in this case are larger than in case of isotropic QD because alignment of individual QD with close interdot spacing causes the system to behave like the elongated QD structure. Nonetheless, its increase is not large since the Stark effect remains weaker than the Coulomb interaction of excitons so that electron and hole behave more like a pair (their wavefunctions are not much changed).

- For the intermediate applied electric field state, it can be clearly seen that increase of electric field intensity builds a high probability of carriers to tunnel outside the well. The effect of electric field separates the electrons and holes to distribute farther away from each other (which is now more dominant than the Coulomb interaction of excitons), but stronger carrier coupling (from the decrease of distance between carriers with an assistance of electric field) produces a larger overlap integral. As a consequence, the polarization degree exhibits a prominent of anisotropy. The maximum polarization degree related to  $V_{PD(max)}$  is represented as the "operating point" of the system. On the contrary, when applied voltage is larger than  $V_{PD(max)}$  (called  $V_{diminish}$ ), the transition probability in the direction perpendicular to the QD alignment (parallel to height of potential well) also increases as carriers in this direction are activated by a larger kinetic energy (from higher intensity of electric field) and by the thermal energy (from phonon corresponding to increase of electric field) as well. This gives rise to decrease of overlapping of carrier wavefunctions, and reduction in the polarization degree.

- In the circumstance of strong applied electric field, sharp decrease of PD takes place. This mechanism is related to  $V_{breakdown}$  which brings the transition probability in both directions to become almost equal. Consequently, this overlarge intensity of electric field leads to a very small amount of PD. Moreover, the higher number of QDs not only makes the degree of polarization to increase, but also leads to a shift of  $V_{PD(max)}$  to a lower voltage (reduces  $V_{threshold}$  too) which implies lower strength of applied field. However, it also creates sharp decreasing of PD value than in case of low number of QDs when a strong field is applied owing to multiplication of the transition probability in the direction perpendicular to the QD alignment and repulsive interaction from the same type of charged carriers.

The electric-field-dependent PD manifests its decrease with increased interdot spacing. Consequently, a large distance between dots causes the shift of  $V_{PD(max)}$  to lower voltage (and produces lower maximum PD value compared with PD in case of small interdot spacing). This is attributed to lower communication of

carriers (less coupling between carriers) from the adjacent dots in the aligned QD direction, indicating the lower of overlap integral. Since overlap integral is significant for determining the polarization anisotropy of aligned QD system, an applied electric field is not only insufficient to increase overlap integral, but also makes it to decrease rapidly at the presence of strong electric field (but nearly unchanged as interdot spacing is overlarge, approaching the case of PD in absence of applied field), resulting in decrease of polarization, as well.

Even though the extending of QD size produces a higher polarization degree under the applied electric field, the results in case of aligned QDs reveal different polarization characteristics. In case of small interdot spacing, it may be clearly seen for higher number of QDs that polarization degree decreases with increasing QD size, which is more obviously seen when a strong field is applied. Focusing on the  $V_{PD(\max)}$ , the shift of  $V_{PD(\max)}$  to lower voltage is observed when the QD size or the number of QDs increases. The PD results in case of larger interdot spacing are almost similar to the case of smaller interdot spacing, and the shift of  $V_{PD(\max)}$  appears at the lower applied electric field. In aspect of physical interpretation, the large dimension of QDs reduces the overlapping between carrier wavefunctions due to strong localization, which is greater than the effect of increasing PD from applied field. In other words, the higher number of QDs with large size, the smaller coupling between QDs therefore occurs, which leads to smaller polarization degree. Furthermore, enlargement of QD size under “strong” electric field affects on the electrons and holes to widely separate from each other so that the optical responses greatly decrease (larger radiative lifetimes), which limits the increase of PD for large QD size.

5. Finally, the interdot spacing in aligned QD system is the “No. 1” factor which influences on the optical anisotropy. It is closely related with the variation of overlap integral. Therefore, the effect of interdot spacing needs to be taken into consideration first, and should be determined properly (as close as possible) to give strong carrier interaction for the high polarization of QD system.

The overall important results as well as the physical explanations have been successfully discussed. The final conclusion of thesis work will be revealed, before pleasingly ending, in the last chapter.