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Coupled quantum wells as a novel source of optical anisotropies in unperturbed nanostructured systems

Doctoral Thesis in Applied Sciences
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Statement of authorship

I, Oscar Ruiz Cigarrillo, student of the Graduate Program in Applied Sciences of the School of Sciences of the Universidad Autonoma de San Luis Potosi, as author of the thesis "Optical anisotropy study in coupled quantum wells, a novel source of undisturbed systems", declare that the thesis is an original, unpublished, authentic, personal work, that the corresponding sources have been cited and that in its execution the legal provisions in force that protect the copyright and intellectual and industrial property rights were respected. The ideas, doctrines, results and conclusions I have reached are my absolute responsibility.

Abstract

In the present work, we were able to identify and characterize a new source of IOAs occurring in asymmetric coupled quantum wells ACQWs; namely a reduction of the symmetry from D_{2d} to C_{2v} as imposed by asymmetry along the growth direction. We report on reflectance anisotropy spectroscopy (RAS) of double GaAs quantum wells structures coupled by a thin (< 2 nm) tunneling barrier (CQWs). Two groups of DQWs systems were studied: one where both QWs have the same thickness (SCQWs) and another one where they have different thicknesses (ACQWs). RAS measures the in-plane optical anisotropies (IOAs) arising from the intermixing of the heavy- and light- holes in the valence band when the symmetry of the DQW system is lowered from D_{2d} to C_{2v} . If the CQWS are symmetric, residual IOAs stem from the asymmetry of the QW interfaces; for instance, associated to Ga segregation into the AlGaAs layer during the epitaxial growth process. In the case of an ACWQs with QWs with different thicknesses, the AlGaAs layers (that are sources of anisotropies) are not distributed symmetrically at both sides of the tunneling barrier. Thus, the system losses its inversion symmetry yielding an increase of the RAS strength. The RAS line shapes were compared with reflectance spectra in order to assess the heavy- and light- hole mixing induced by the symmetry breakdown. The energies of the optical transitions were calculated by numerically solving the one-dimensional Schrödinger equation using a finite-differences method. Our results are useful for interpretation of the transitions occurring in both, symmetric and asymmetric CQWs.

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ABBREVIATIONS

BZ	<i>Brillouin zone</i>
QS	Quantum Structures
QW	Quantum Well
SQW	Single Quantum Well
CQWs	Coupled Quantum Wells
VB	Valence Band
CB	Conduction Band
SCQWs	Symmetric coupled quantum wells
ACQWs	Asymmetric coupled quantum wells
RAS	Reflectance Anisotropy Spectroscopy
PL	Photoluminiscense spectroscopy
PR	Photoreflectance spectroscopy
R	Reflectance spectroscopy
PRD	Photo-Reflectance Differential Spectroscopy
FDM	Finite differnce method
CCD	Charge coupled device
0D	Zero-dimensional
1D	One-dimensional
2D	Two-dimensional
3D	Three-dimensional
fcc	Face-centered cubic
2DEG	Two-dimensional electron gas
BL	Beer-Lambert-Law
TB	Tight-Binding method
PD	Photo-Detector
PEM	Photo-Elastic Modulator
QM	Quantum Mechanics
$k \cdot p$	Semiempirical theoretical tool to calculate band-structure
TB	Semiempirical Thight-Binding Method
SOC	Spin-Orbit Coupling, also called Spin-Orbit interaction
LFLM	Dr. Luis Felipe Lastras Martinez group.
EFA	Envelope function Approximation
EMA	Effective Mass Approximation
FKOs	Franz Keldysh oscillations
IOA	In-plane Optical Anisotropy

LIST OF CODES AND PACKAGES

This list denote the *Open-Source* packages, codes, tools, and repositories for the development of this work. All inside of this work as images or numerical calculations are subject to the *Open-Source* ideology. Our codes are housed in our own GitHub repository, both personal and laboratory repository. It's important to say that without the development of the *Open-Source* codes like contents in this list, our codes, they couldn't be enhanced.

cqws-codes Repository of our codes implemented in this work. [1]

kp-lflm-group $k \cdot p$ julia [2] package developed by Our group research [3]

ASE The Atomic Simulation Environment (ASE) is a set of tools and Python modules for setting up, manipulating, running, visualizing and analyzing atomistic simulations. [4]

Spglib Software library for crystal symmetry search [5]

SOLCORE A multi-scale, Python-based library for modelling solar cells and semiconductor materials [6]

Aestimo One-dimensional (1D) self-consistent Schrödinger-Poisson solver for semiconductor heterostructures [7]

VESTA 3D visualization program for structural models, volumetric data such as electron/nuclear densities, and crystal morphologies. [8]

PGF/TikZ PGF is a macro package for creating graphics. It is platform- and format-independent and works together with the most important TeXbackend drivers, including pdfTeXand dvips. It comes with a user-friendly syntax layer called TikZ. [9]

pst-optexp PStricks package to drawing optical experimental setups. [10]

SYMBOLS

X⁻ Negative Trion

X⁺ Positive Trion

X Direct Exciton

IX Indirect Exciton

$\text{Al}_x\text{Ga}_{1-x}\text{As}$ AlGaAs semiconductor as a function of Al concentration x

\hbar Planck's constant (eV)

m_0 electron effective mass

(hkl) Family of lattice planes with Miller indices h , k and l

E_g Energy bandgap

e electron

hh heavy-hole

lh light-hole

$e_n - hh_n$ **or** $e_n - lh_n$ Electronic transitions

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1

PHYSICAL BACKGROUND

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QUANTUM MECHANICS in semiconductors is basically the electron behavior that exhibits many phenomena non explained by a classical regime. Quantum structures (QS) are artificially systems conformed by semiconductors where electrons exhibit their quantum nature, which is a great platform to study and create quantum devices. Nowadays, the progress in creation of QS consist in precisely deposition of thin films, in which electrons show fundamentally new electrical and optical properties [30]. Most of these properties show energy confinement, which is the principal interest to study the electrons and their consequent interactions which generates analogous hydrogen atom in semiconductors. Therefore, the interest to study QS was increasing for many years ago and nowadays, these continue considering an excellent research area.

In this chapter, it is presents the fundamental concepts to describe the physical phenomena reported in this work, without intention to replicate concepts and models already explained in publications with major impact. Therefore, the purpose is to present to own interpretation to highlight the importace the breakdown of symmetry in semiconductor QWs.

1.1 Semiconductor Bandstructure

We start with the understanding the band structure of semiconductors. The band structure describes the electron behavior in a solid; therefore, we will need to invoke the Scrödinger equation. Inside a solid, around 10^{23} valence electrons contribute to the bonding in each cubic centimeter, which results in a many-body complex problem [31], with a general hamiltonian [32, 33]:

$$H = \frac{1}{2M} \sum_{i=1}^{N_n} \mathbf{P}_j^2 + \frac{1}{2m_0} \sum_{j=1}^{N_e} \mathbf{p}_j^2 + \frac{Z^2}{2} \sum_{i,j=1, i \neq j}^{N_n} V_c(\mathbf{R}_i - \mathbf{R}_j) - Z \sum_{i=1}^{N_n} \sum_{j=1}^{N_e} V_c(\mathbf{r}_j - \mathbf{R}_i) + \frac{1}{2} \sum_{i,j=1, i \neq j}^{N_e} V_c(\mathbf{r}_i - \mathbf{r}_j), \quad (1.1)$$

where N_n is the number of atomic nuclei, N_e is the number of electrons with mass m_0 , asumming that the nuclei are the same mass it's consider M and charge Z_e . As is obviously, this Hamiltonian is so complicated, the sum of five terms which consists in : kinetic energies to electrons and nuclei, the nucleus-nucleus, nucleus-electron and electron-electron Coulomb interactions, also \mathbf{R}_i are the positions of the nuclei and \mathbf{r}_j are the position of the electrons, the operators \mathbf{P} and \mathbf{p} are momentum operators to nuclei and electrons respectively. Finally, the Coulomb potential V_c [32]is also considered.

Fortunately, the QS are formed by crystalline materials the Bloch theorem provides the most important tool to develop required equations. The Bloch theorem establishes for a periodic potential $U(\mathbf{r})$ for electrons, due to the material's periodicity (definition of crystal structure) the Schrödinger equation can describe in terms of "single electron" is given by:

$$\left[-\frac{\hbar^2}{2m_0} \nabla^2 + U(\mathbf{r}) \right] \psi(r) = E\psi(\mathbf{r}). \quad (1.2)$$

The principal reason that the periodic potential in a crystal structure is highly important is their translational invariance concept and the consequent symmetry operations that are possible in a crystalline solid. The symmetry concept, as a tool to understand solids, is discussed with major detail in the next chapter. In according to Bloch's theorem we can associate a wave vector \mathbf{k} with each energy state, $E_n(\mathbf{k})$. Thus, it is useful to display the energies $E_n(\mathbf{k})$ as a function of the wave vector \mathbf{k} . This is also known as dispersion relation, but in general terms is the electron band structure of the given solid [31].

Even if the calculation of electron band structure in solids is a very complex task due to the distance between atoms, the Bloch's theorem provides the most important tool to reduce the problem of crystalline structures. For this there exists, several methods to calculate the realistic bandstructure for semiconductors that are categorized in two groups: Atomistic methods* (Tight-binding, orthogonalized plane wave methods) and Perturbative methods $(\mathbf{k} \cdot \mathbf{p})^\dagger$. These two main categories with theirs respective methods have special characteristics which becomes in the reasons the best chooice. These reasons, have to do with the description of bandstructure. Therefore, each of these methods can be chosen and enhanced as the system to study requires. We won't enter in discussion about which of these methods are the best as each method is powerful, and we must to keep in mind that the complexity of solutions requires to be solved by numerical techniques with very good approximations. Then, basically the electron behavior inside semiconductors consisting of solutions for the appropriate Schrödinger equation [34].

1.1.1 Valence and Conduction Bands

The most important characteristic of semiconductors is the bands structure. In general, the bands of semiconductors composed by valence and conduction bands separated by a region known as bandgap. The bandgap, which is proportional to separation energy of valence and conduction bands, also is called as forbidden region. This is because does not exist electron states inside; therefore this gap energy determine the electron conduction in

*In this category can include the ab initio methods, these are the most complex methods due to propose solutions of the many-body problem

[†]In fact, both TB and $\mathbf{k} \cdot \mathbf{p}$ also consider in same kind, because both are *semi-empirical* methods due to they consider experimental parameters.

a semiconductor and this is the difference they exhibit with the insulators and metals as shown Figure 1.1.

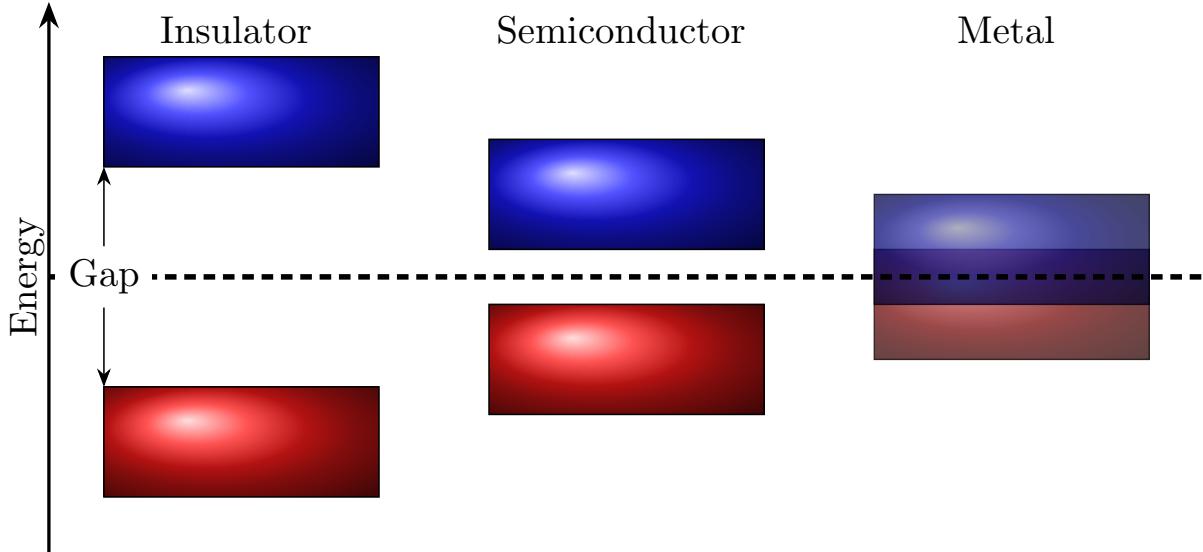


Figure 1.1: Band energy diagram for insulators (left), semiconductors (center) and metals (right). The principal difference is the gap energy: for insulators, this is bigger than semiconductors, . In metals there is not a definite gap energy, and instead there exist an overlap of bands. Dashed line determine the Fermi level.

The bandgap determines many characteristics and functionalities in semiconductors. Moreover, the bandgap energy classifies semiconductors in direct and indirect semiconductors. As pointed out before, it is so difficult to describe electrons behaviors inside solids due to the many body interactions that exists, and therefore the Schrödinger equation that's describe electrons behaviors is a unrateable task. Fortunately, the semiconductor structures have a one of the most important characterize and it has to do with their atomic structure: it's periodically arrangement of atoms. This periodicity is key to propose solutions and describe the semiconductor band structure. Starting with the description bulk semiconductors, for example GaAs consisting of III-V atoms with cube simmetry, that can be thought as to inter-penetated FCC sublattices, as shown in Figure 1.2. For this case, when the atoms in two sublattice are different, the crystal structure is then called *zinc-blende* [35].

To calculate bandstructures of bulk semiconductors it's important to define specific symmetry direction, this mean that it's not possible to plot dispersion relation. For each three-dimensional wave vector \mathbf{k} , then the plot energy as a function of \mathbf{k} is along of different high-symmetry directions [31]. In this work the structures to study are composed of semiconductors III-V, being GaAs the bulk in each structure. GaAs is a direct semiconductor so that the [001] direction is the high-symetry direction then is denoted by Γ point (at $\mathbf{k} = 0$)

As before mentioned several times, it's very complex to compute the Schrödinger equation in the solids. The most “exact” compute is employed by DFT theory, these calculations

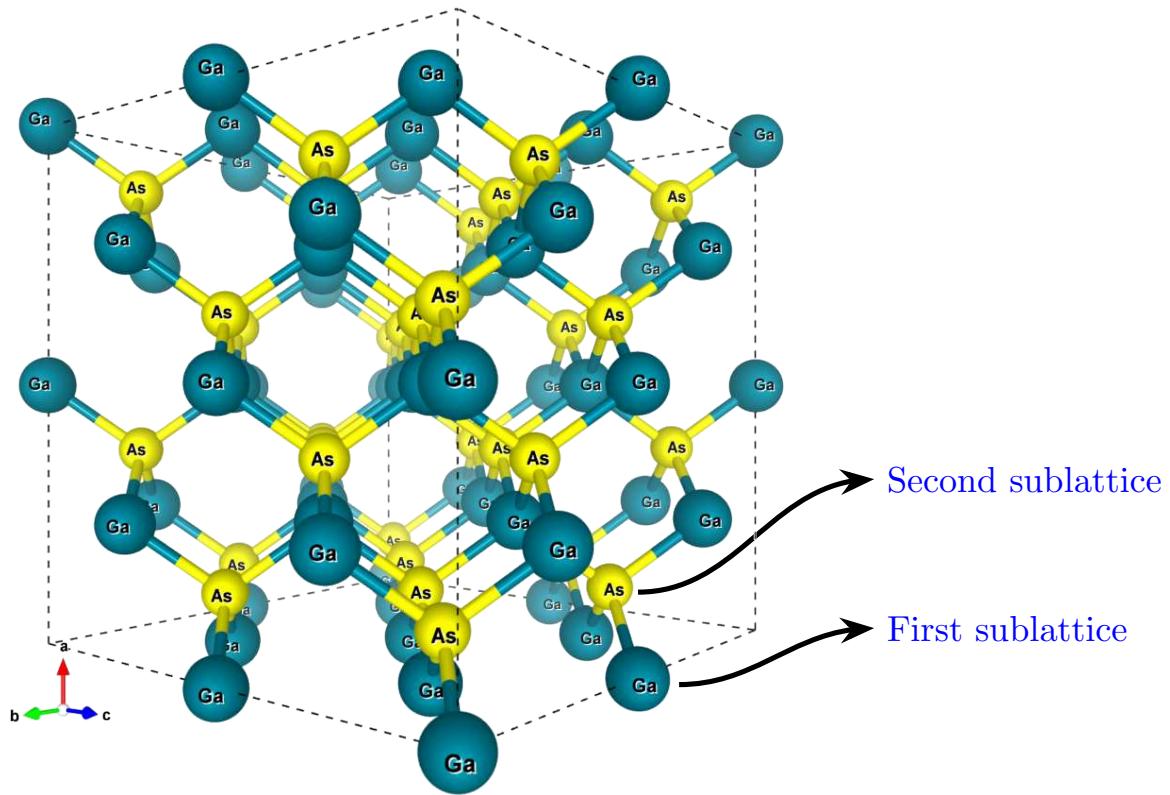


Figure 1.2: GaAs crystal lattice, where each sublattice corresponds to each atom species Ga and As.

commonly are called atomistic even some semiempirical models can consider as atomistic, but the semiempirical models are good approximations in comparison with the DFT theory. So, which is the reason to call “exact” solutions to the DFT results? The answer leads us to great discussion and it’s not intended to get into controversy, but in general the DFT calculations have the capacity to calculate in terms of electrons interaction and the empirical methods are based in potential choice.

We will, don’t go into details about band calculations theories and models, but we will make a general reference to the importance in this work. The models more performed in semiconductor heterostructures as GaAs/AlGaAs are semiempirical, this is because DFT theory and their derived models are very limited to carried out in large structures, their electron interaction nature need high computational power. So that in comparison with empirical models where the main role is the potential of semiconductor structures, this reduces computational reduce. So, the most models used in semiconductor band calculations are empirical models, these models are distinguished by low computational requirements for this reason are considered like approximations. The importance to discuss these concepts will take relevance when we discuss the physics model proposed in this work.

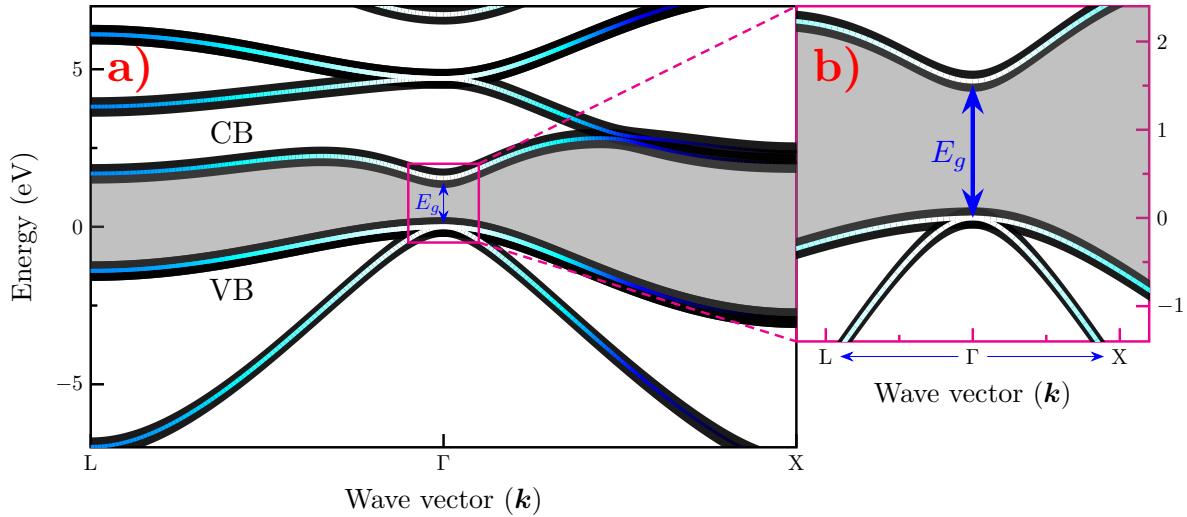


Figure 1.3: Band structure of GaAs, (a) shows the zoom around of Γ to denote the direct band gap and the electrons energy needed to jump from valence to conduction band. (b) denotes the two directions to dispersion of the bands corresponds to Brillouin zone: $\Gamma \rightarrow X$ and $\Gamma \rightarrow L$. [11]

The Section 1.1.1 shows the results of calculations of TB model as discuss it in [36] and the code was implemented by R. Muller [37]. The model purposes by Vogl et al. take into account small number of localized pseudo-orbitals and based the empirical parameters to substitute on TB Hamiltonian. The importance to get bandstructure it's based importance to study optical properties of solid structures, if it doesn't exist band electrons it's like look a place without map, so, the band structure far from being a complex tool it's the key to get the information to investigate the optical properties.

As shown in Figure 1.3(a) the GaAs bandstructure shows that it's a direct semiconductor as a previously mentioned, this gives way to get electron transitions from VB to CB and the energy to success this it. In Figure 1.3(b) it's plotted, the band dispersion around Γ point, it's the most symmetry point. It is well-known that the band dispersion increasing \mathbf{k} along two different directions of the Brillouin zone, from $\mathbf{k} = (0, 0, 0)$ to X point $\mathbf{k} = (2\pi/a_L)(1, 0, 0)$ and L point $\mathbf{k} = (2\pi/a_L)(1, 1, 1)$. So, these figures, are the typical representation of direct gap to III-V semiconductors around of $\mathbf{k} = 0$, then, is obviously that the shape of dispersion is parabolic. For the GaAs bandstructure calculations shown in Section 1.1.1 doesn't take into account the contribution of spin*, therefore it's focus on three bands dispersion correspond to a single *s*-like conduction band and two *p*-like valence bands. Is important to say that the characteristic curvature $E-\mathbf{k}$ of dispersion bands corresponds to an electron (*e*) in case of positive curvature while the negative curvature correspond to holes states; heavy (*hh*) and light hole (*lh*) bands, so, this denotes that the transitions, are of dipole nature [11, 33]. All the above disputed is significant to refers of one of the most indispensable quantum mechanisms in solids, this is absorption. The

*This spin contribution as called as split-off (so) hole band.

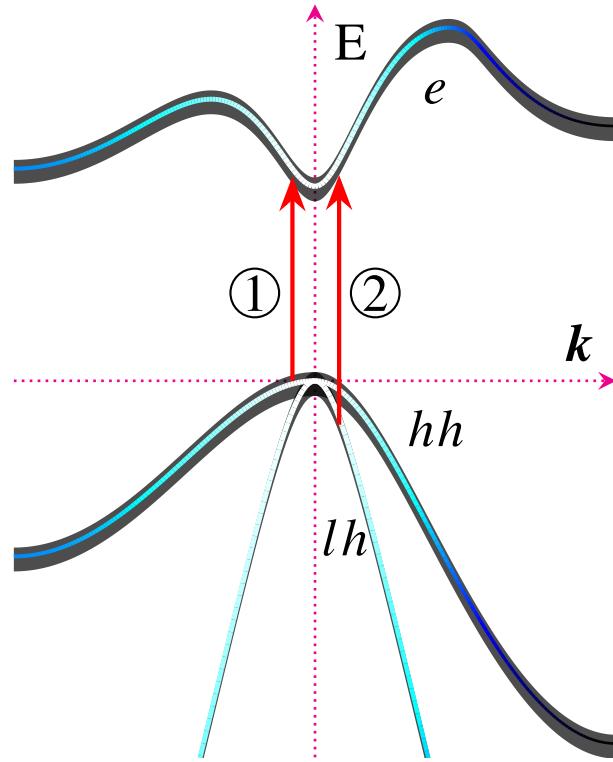


Figure 1.4: Two typical transitions for GaAs near $k = 0$. The first one correspond to the heavy-hole and second one to the light-hole.

electron absorption, specifically interband absorption, give way to a fundamental physical process that involves the principle of many basic studies of semiconductors, applications, and the importance to understand the electron behavior in a semiconductor structures disputed in this work. Then the Figure 1.4 schematizes the two typical transitions in GaAs bulk, this transitions are near to $k=0$, so that is called interband absorption. Is important to remark that the interband transitions are observed in all solids, but the mechanisms are different dependently of their bandstructure, for what, being repetitive when mentioning that the bandstructure is the key to study solids.

In case of GaAs the interband transitions are called as direct transitions, this is because their bandstructure proofs GaAs is a direct Gap semiconductor. This process is determined by quantum mechanical rate $W_{i \rightarrow f}$ for exciting an electron in a initial quantum state ψ_i to final state ψ_f by absorption of a photon of angular frequency ω [11]. As is very known, this is given by Fermi's golden rule. Later this is disputed according to highlight the model and results obtained. It has been mentioned that the direct transitions are of dipole nature, as before mentioned the CB is type *s*-like while the VB is *p*-like, then it's electric-dipole allowed transitions $p \rightarrow s$. The excitation of electron in CB carries to leave an initial state unoccupied, this is called as a hole creation, then the electron in the final state and the hole is considered as **electron-hole pair**. The next part subject this theme with major focus.

1.1.2 Excitons

The importance to study bandstructure of semiconductors is very clear so far, so that could be said that absorption process is the source of optical properties of solids. It's due to this that the importance to study of semiconductors in the optoelectronics applications. But, this process give rise to formation of one of the most important excitations in the crystal structures. The photon absorption process carries to an electron is excited from CB to VB, this generates an empty location in VB which has positive charge. This positive empty location called as hole, therefore the electron and hole have opposite charge then it's to be expected that they are attracted, so, this creates a bound state called an exciton [38].

From Figure 1.4 and Figure 1.5 is clearly that excitons are commonly presented in direct band gap semiconductors as GaAs, this was denoted by absorption experiments, after mentioned in Section 3.2.1 this is the cause in photoluminescence mechanism.

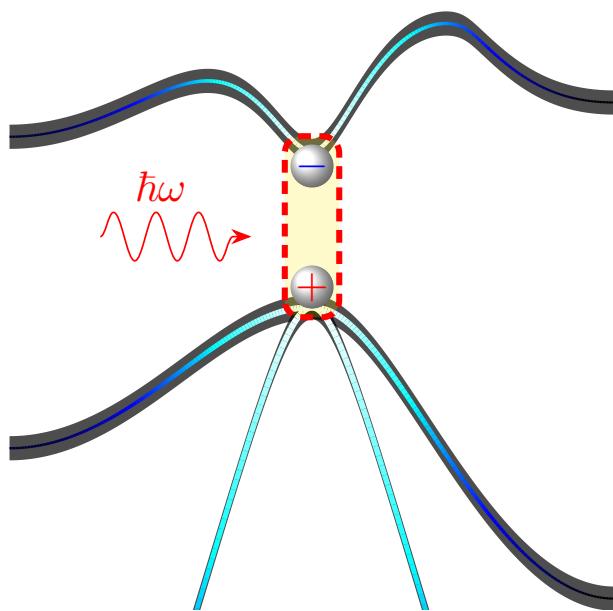


Figure 1.5: Qualitative scheme of exciton creation in GaAs as direct gap.

1.2

Semiconductor Low-Dimensional Structures

The previous section engaged to explain the principles of semiconductors, this is the bandstructure, the importance of these is practically the fingerprint of all semiconductor, without bandstructure the understanding of these would be improbable. The first approximations were based in GaAs bulk, their cubic symmetry practically defines their nature and consequently the physical effects as excitons existence. But, what happens if joined several semiconductors with same symmetry and structural parameters? The bulk properties and physical properties are the same?. The answers they are well-known, when two materials with relatively same structural parameters, as lattice constant can create a heterojunction, the union of several heterojunction make up a heterostructure.*

The Figure 1.6 is a general scheme of a heterostructure, in this case is presents three

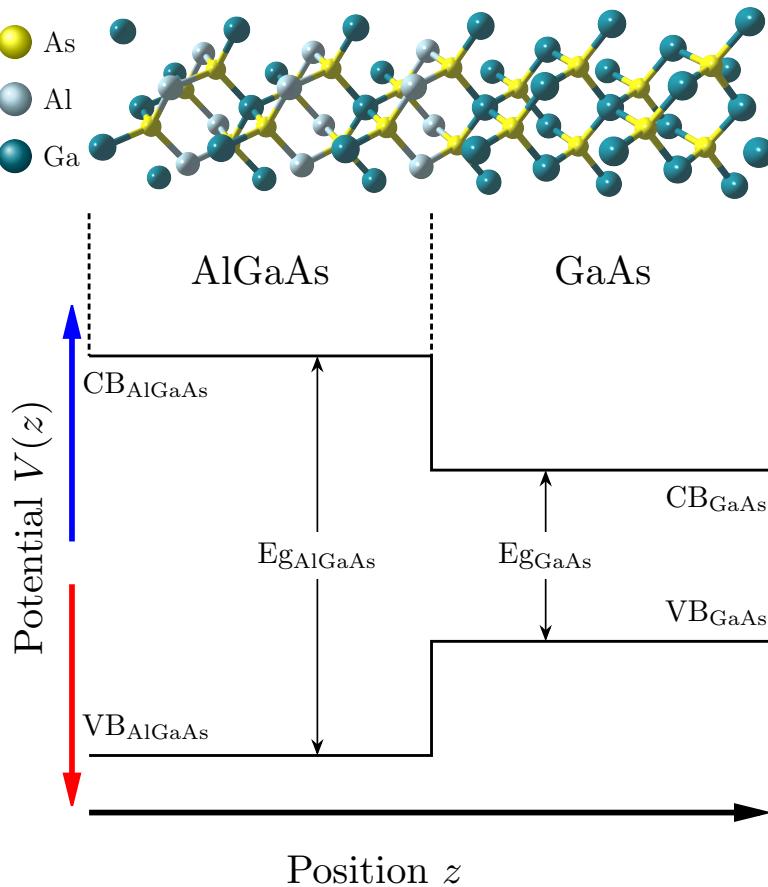


Figure 1.6: General scheme of $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterostructure, at top show the scheme of atomic arranged of this heterjunction, the dashed lines are the matched between two dissimilar materials. In bottom shows the band-edge profile.

*The samples studied in this work are heterostructures, for this reason, and by nomenclature it's we refer to that way.

species of atoms Al, As, Ga. These atoms can locate in columns III-V of the periodic table, hence its name of III-V semiconductors. The principal characteristic of these atoms is that it can create matched structures as GaAs, AlAs and ternary alloys as $\text{Al}_x\text{Ga}_{1-x}\text{As}$ with specific Al concentration. The matched semiconductors produce a material with new properties based principally in the difference of bandgap which involves the alloys.

As can see in Figure 1.6 it's consisting a GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterostructure, these interface is well-matched due to the lattice parameters is relatively equals, therefore and thanks to powerful growth technics as MBE it's possible to get high-quality quantum structures.

Also, the heterostructure composed by two semiconductors with different band gaps generate a discontinuity in either the conduction or the valence band can be represented by a constant potential term [39]. The theory to treatment the electron behavior in these structures, is relatively simple if we consider the above. Although, in this chapter doesn't have intention to explore the theory of electron behavior in that, worth noting that it get one-dimensional potential $V(z)$ to both bands, so the Schrödinger equation can solve simple.

1.2.1 Quantum wells

Doubtless the creation or growth of heterostructures increased the interest in the study of quantum structures, the interactions, and physical behavior of light-matter they would not have been possible without these. The major relevance is due the quantum confinement, the junction of semiconductors results in an interest quantum structures with specific dimensions. From 3D bulk the dimensions reduce to 2D, 1D and 0D dimensional structures. Therefore each of that has interest properties and their correspond applications. The Figure 1.7 schematics the low dimensional heterostructures from 3D bulk, the first low dimensional from 3D to 2D is the Quantum Wells, then from 2D to 1D it have the Quantum Wires finally with 0D have the Quantum Dots. The quantum confinement so is the principal reason to study that structures, the electron behavior which exhibits in it should can to help understand a great variety of quantum mechanical phenomena as electron interaction on a crystal. Suppose a heterostructure composed with a two semiconductor alloys as sandwich, this 2D quantum structure is called a Single Quantum Well (SQW).

This dissimilar semiconductors in terms of their potential energy ($V(z)$) can be schematized as Figure 1.8. The Gap difference of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ and GaAs is due to x Al concentration in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ therefore it obtains a one dimensional potential profile, with that can confinement electrons in a 2D plane along z direction. All of these carries to quantum mechanics formalism, the electron behavior should be obeyed these rules. If we have an electron closed in two potential barriers an L distance, the wave which describe it will be spatially confined. So if we confined many electrons in these potential, we have two important physical aspects: the first one is knowing as Pauli's exclusion principle,

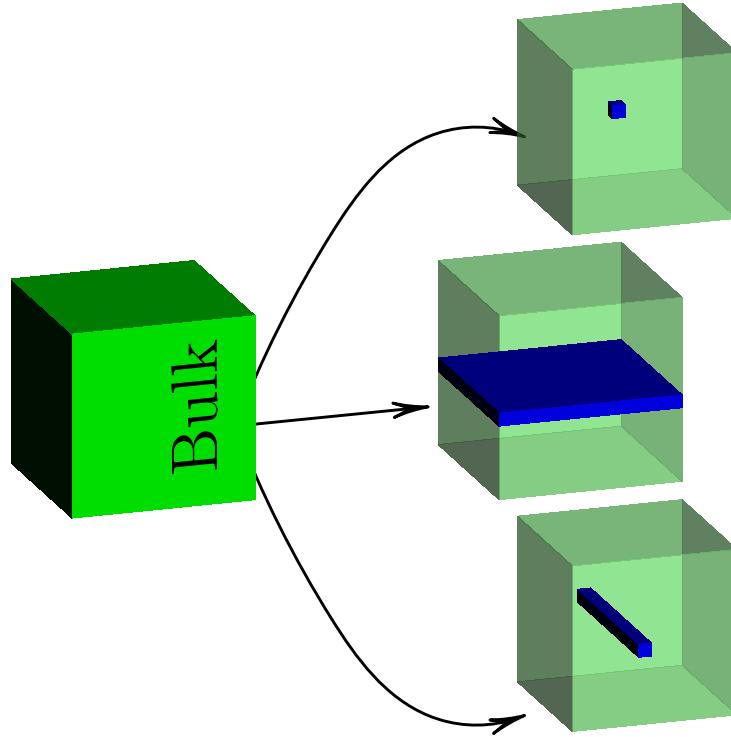


Figure 1.7: Heterostructures from bulk (3D), to Quantum Wells (2D), Quantum Wires (1D) and Quantum Dots (0D).

which as of its Fermion nature prevents carriers with the same spin occupying the same region in of space [39, 40], the second one and one of the most relevant in the birth of the quantum mechanics; the Heisenberg's uncertainty principle. That last, it can say that is the consequence of quantum confinement due to the space reduction of the electrons is expected that momentum increases by an amount of the order \hbar/L . Therefore, the energy of that confined particles increases, and it's referred to as confinement energy [33].

Then the quantum confinement is our started point to understand the optical properties in QWs. As is referred in the figure, the uni-dimensional potential profile can well describe by top conduction- and bottom valence-bands, the band offset in these two of correspond gap energy between that, while Al concentration increases their bandgap and the band offset (Q_c to CB and Q_v to VB) also to.

It's so clearly that the QWs have the potential to presents amazing quantum properties, even if all of these are very important we focus on the optical properties, basically our interest is the light-matter interaction through its result mechanisms.

1.2.2 Preliminary approach of Quantum Confinement effect in QWs

As the title describes, here it will try to explain as the first approach the quantum confinement effect in QWs. If it starts with the scoop, which it can be reduced the

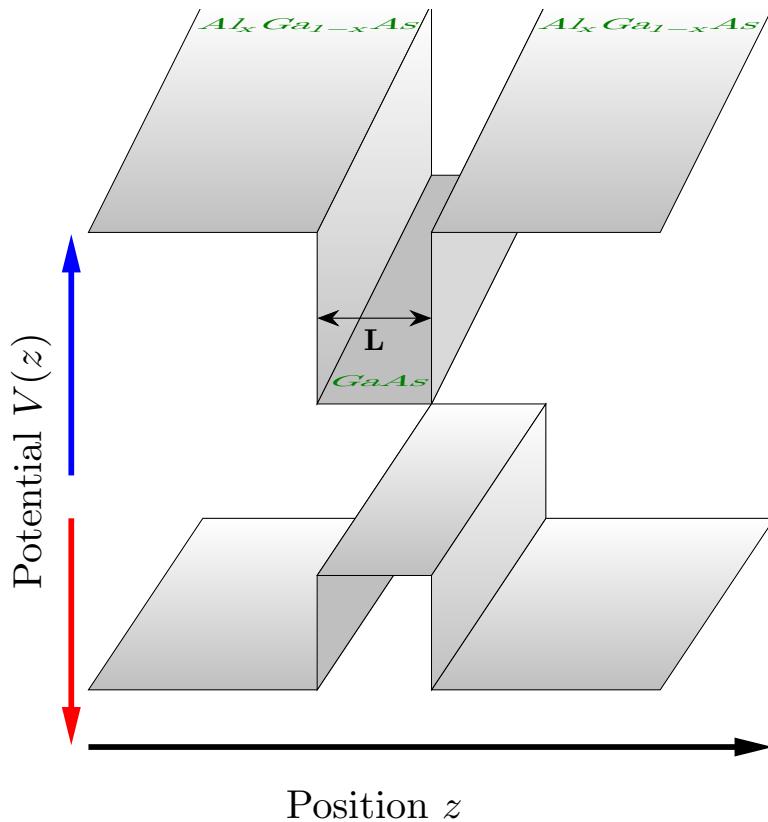


Figure 1.8: $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ Single Quantum Well

electrons space, then this mean that in reciprocal space it has two components k_x and k_y . Then say in crystal symmetry properties to the case of $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ QWs it's Γ the central point, as long as $x < 0.4^*$ the bandstructure depends on confinement energ, so say which the bandstructure depends on confinement energy. In this case, the electronic properties in comparison with a bulk semiconductor properties can solve trough particle-in-a-box as textbook problem as first approach. Nevertheless, even if usually can solver without much mathematical formalism is very essential that it dedicates a chapter with their solution, this is because will employ a physical formalism exclusively to QWs structures. In general way, as it before mentioned the Schrödinger equation solution is the fundamental pillar to understand, where it's taken into account which in a crystal the periodic potential is the key. Here are important remarks before to continue, when it has a heterostructure starting with the bulk model it's clearly that the system doesn't same, the Quantum Mechanics which is behind take into account the symmetry properties, then it can be developed a Hamiltonian to understand that system. In the next chapter will be explained details and the formalism both physical and mathematical to solve and discuss it's. The model which give the tools to get the solutions is called as Effective Mass

*It will be explained in the next section, although it's due to the Gap go from direct to indirect, shortly the symmetry $\Gamma \rightarrow X$.

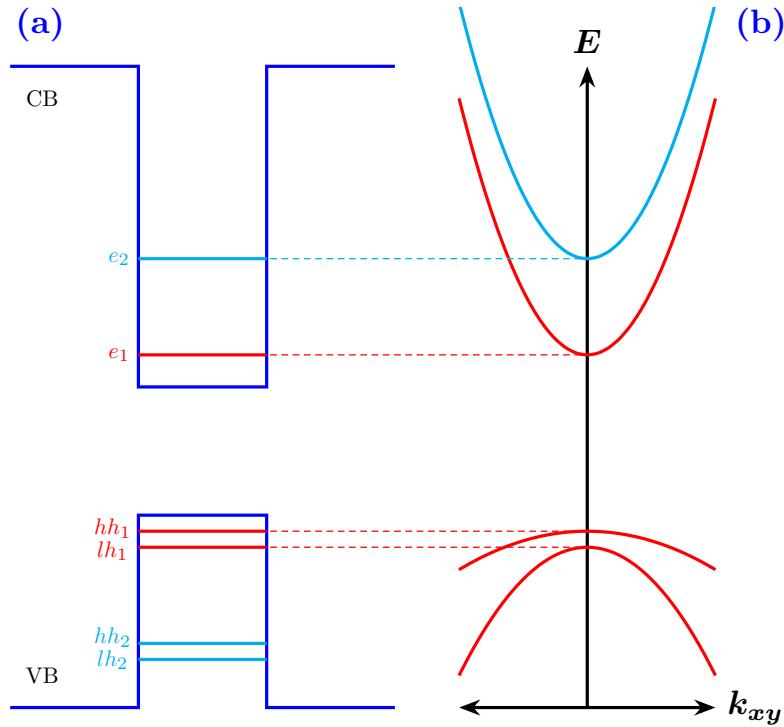


Figure 1.9: General scheme of typical Schrödinger's equation solutions to one-dimensional potential as (a) where the Eigenenergies of both electron and holes are denoted with same color depending on n value.(b) It's plot, of the subbands in the same case of (a) to both particles.

Approximation, thus their correspond Schrödinger equation is [11, 39, 41–44]:

$$-\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial z^2} \psi(z) + V(z)\psi(z) = E\psi(z), \quad (1.3)$$

where the m^* is the effective mass in each material, and $V(z)$ is the potential profile got by heterostructure materials properties. Therefore, that differential equation can solve as in textbooks explained [11, 39, 41, 43, 45–48]. The idea is thinking as a one particle in a finite potential well, where is well important established the boundary conditions and solve the Schrödinger equation in each part of single QW, this means that need to create a potential function. Then it's can obtain the Eigenfunctions and their correspond Eigenenergies.

The principal idea doesn't is reproducing something which is very well known, the objective of this part is established the scoop of the next chapter. Therefore, before to continue, we will finish with to explain the dispersion in-plane of single QW. As in the QW the one-dimensional potential set up the 1D confinement, is important doesn't confuse which the QW is a 2D structure, but their confinement is along of z direction this mean 1D. Then, the particle can motion in the $x - y$ plane. By this reason, even if consider 3D Schrödinger equation and the above is considered it obtain Equation (1.3) therefore, the solutions in the one-dimensional potential produce discrete states of energy $E_z = E_n$ [39],

where n is the energy level it which produce subbands as shows in Figure 1.9. In contrast, before it called as “energy bands” in the bulk case, now due to the quantum confinement gets subbands to both conduction- and valence bands.

These subbands are the result of the sum of E_z and $E_{x,y}$, which are the 1D confinement energy and the in-plane momentum $k_{x,y}$ then [39]:

$$E = E_n + \frac{\hbar^2 |\mathbf{k}_{x,y}|^2}{2m^*}. \quad (1.4)$$

From equation the effective mass m^* depends on particle, i.e the effective mass to electrons in CB and the holes in VB. So, the most relevant in the solutions is the energy E_n (Figure 1.9(a)) is discrete, this is the quantum confinement in the low-dimensional heterostructures.

1.3 Summary

In this chapter, was exposed the generalities of semiconductor band structure and low-dimensional heterostructures, highlighting or taking in major GaAs/Al_xGa_{1-x}As that's the semiconductors of major importance in this work. The band structure interpretation usually be so hard, and their calculations even more, but the impact and relevance in optical properties of semiconductors starts from that interpretation, from here arises the mathematical arsenal to right physical interpretation. Another significant concept which was treatment as first approach is the effective mass concept, even if, when solved, the bulk Hamiltonian it considers the mass as constant parameter or depending on semiconductor material, contrary in low-dimensional structures have an important role.

In generally, the band structure of semiconductors is the key to understand quantum properties of solids, in this work the relevant is the light-matter. Remember that light-mater interaction in solids can be studied by process resulting in it, as absorption, reflection, transmission, diffraction, scattering, and others [49]. Although, the light-matter interactions are fundamentally quantum electrodynamical, also, can be studied in quantum way through before mentioned process. Firstly, is the photon absorption process can help to understand or calculate the fundamental parameter in semiconductors as is bandgap energy. This parameter is the started point in the study of semiconductors, this is the start point in the map called bandstructure, if it ignores the gap value in the semiconductor to study it couldn't possibly get the principal optical properties of that.

Then, the bandstructure of semiconductors is the map to understand them, without these routes or fundamental parameters couldn't have quantum devices.



PHYSICAL MODEL

In this chapter, it's exposing the fundamental physics to understand the experimental results' through physical model, both numerical and phenomenological. It is emphasis in symmetry properties of semiconductors, which is the base to get our model.

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SEMICONDUCTORS are alloy of materials with pure structural characteristics, it's the alloys which can generate an amazing quantum process that is quantum confinement. But, all of them couldn't possibly be achieved without understand of semiconductor bands, are bands the reason to get semiconductor heterostructures. Now, the objective is proposing the model to study Coupled Quantum Wells. The CQWs are heterostructures grown from a semiconductor substrate as GaAs as previously treated SQW (see Figure 1.8) but coupled by a thin barrier which have a very significant role. But before to mention the objective of study this structures and the physical model which explain their experimental results is important to call about of the symmetry and their relevance to understand the physics of these QS.

Previously, we were very repeating in the importance of semiconductor band structure, also to remark difficult of get them. It was saying the complexity of calculate semiconductor bands is high, by this reason it's developed models to approximate it. But, all of these couldn't be possible if it would not be taken into account the *symmetry* role in physics [50]. Thanks to symmetry, it's possible to understand from Quantum to Universe. It is clear that to talk the symmetry is inevitable to think in geometry or nature patterns inherently way, but in the next sections it's established the symmetry role from the band structure calculation to the importance in the electron behavior in CQWs structures.

2.1

The Symmetry Context

Talk about *symmetry* is talk of shapes or in a romanticism way as the natural harmony that makes something appear beautiful to us [51, 52]. But, what is the reason the *symmetry* is very important in physics?. The reason the *symmetry* is very important has to do with a *transformation*, this mean that if a physical system is affected or perturbed by a thing and this appears to be exactly the same before and after that *transformation*, it is said to be *invariant* under that *transformation*. The symmetry of the system is made up of all the transformation operations that leave the system invariant [51].

In this section doesn't have the purpose to be a one more copy or re-interpreted version of a group theory book, yes, the group theory not symmetry theory, it's important to remember that to understand of symmetry physics of solids it's important to understand the group concept. In generally, a group is a set or collection of elements that obey certain criteria and are related to each other through a specific rule of interaction and obey four group axioms [51, 53, 54]. It is important to remark which the heterostructure to will study is composed by GaAs/Al_xGa_{1-x}As semiconductors, then we started with the GaAs crystal to propound the symmetry role in the CQWs. Then, already raised the starting point, remember that the crystal solid can be defined as an arrangement of atoms in strictly periodic arrays [55, 56], from here arises two concepts: basis and lattice, where that

last is the set of mathematical points to which the basis is attached [55]. These crystal concepts give place of crystal primitive cell in three dimensions also considered as the seed to reproduce a crystal. So, it gets fundamental types of lattices defined by a collection of *symmetry* operations (rotation, translation etc.), then it's compose a lattice point group. In the three-dimensional case, the point symmetry groups require the 14 different lattices types*, where are classified into seven systems. Into these systems it's found the *cubic* system, it which posses three number of lattices. Remember that the GaAs crystal is *cubic*, specifically, is the type FCC lattice. The FCC lattice is easy to imagine, if place an atom in each corner of a cube and in a center of each face of it. Therefore, it is easier to define the planes and crystal directions if we take the cube faces as a reference it gets (hkl) plane and the directions [hkl] which must be perpendicular to a plane (hkl) [55].

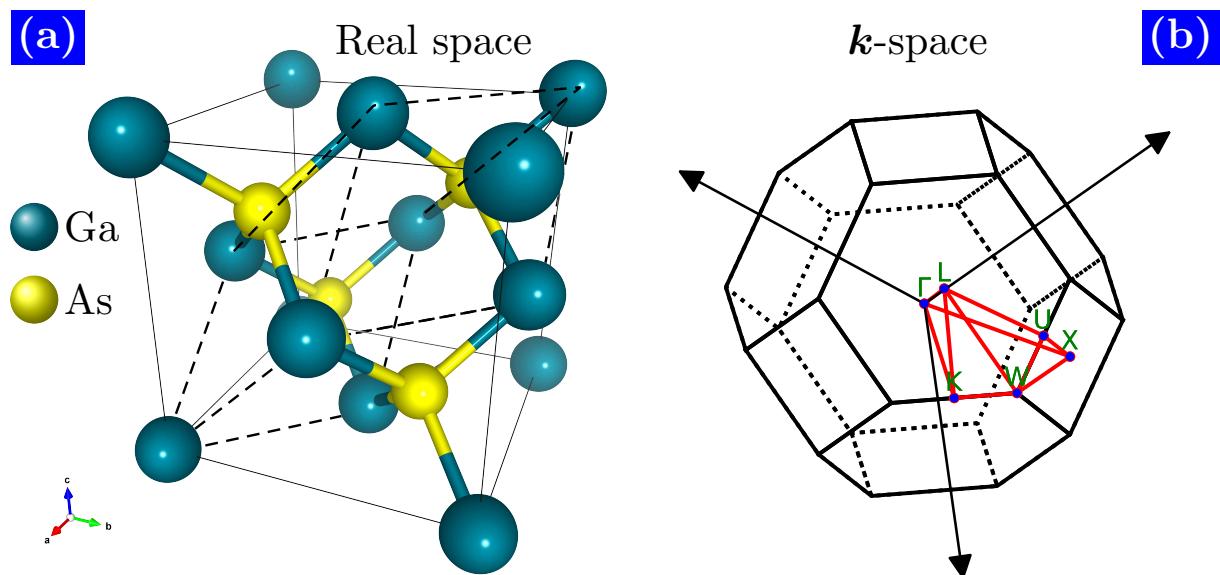


Figure 2.1: (a) GaAs crystal structure in “real space”, this region is known as *unit cell*, into that with dashed line is denoted the primitive cell. This lattice is well-defined by the vectors \mathbf{a} , \mathbf{b} and \mathbf{c} , these vectors are defined as the basis vectors. In (b) is schematized the GaAs crystal structure in k -space, also known as *reciprocal space*.

The lattice is an array of points which make the space lattice of a *crystal* and the repetitions or disposition of these points is controlled by “*symmetry* operations” [57]. The crystal is composed by a space lattice this is a plane lattice[†] which have three *symmetry* operations: *Rotation axes*, *Mirror plane* and *Centre of Symmetry*. If add a one dimension to plane lattice it gets a space lattice, which define the unit cell of a *crystal*, so this adds one more *symmetry* operation which is *Rotation Inversion* or *Roto-Inversion*. Then, can get *symmetry* elements of a *crystal* if apply the four *symmetry* operations and their possible combinations. If collect that *symmetry* elements obtains the *point symmetry* or the *point*

*This lattices are known as Bravais Lattices

[†]2D point pattern array

group of symmetry of a *crystal*. The GaAs crystal as before mentioned is a *cubic* system, but have a defined *cubic* structure called as *cubic zinc sulfide* or simply *zincblende*. This specific *cubic* structure is characterized by arrangement of two type atoms with places coordinates: $000, 0\frac{1}{2}\frac{1}{2}, \frac{1}{2}0\frac{1}{2}, \frac{1}{2}\frac{1}{2}0$ for one type of these as Zn in ZnS or Ga in GaAs structure. In case of the second one atom, it has coordinates : $\frac{1}{4}, \frac{1}{4}, \frac{1}{4}, \frac{1}{4}, \frac{3}{4}, \frac{3}{4}, \frac{3}{4}, \frac{1}{4}, \frac{3}{4}, \frac{3}{4}, \frac{1}{4}$ for S in ZnS or As in GaAs [55, 58]. The Figure 2.1(a) shows the unit cell of GaAs structure and into them it dashed the primitive cell for the FCC lattice, also denoted the basis vectors \mathbf{a} , \mathbf{b} and \mathbf{c} .

In the case of symmetry of GaAs crystal, is important to remark that this symmetry can also denote in Hermann-Maguin notation $F\bar{4}3m$ which corresponds to three fourfold rotary inversion parallel to the edges of a cube, with four threefold rotation axes parallel to the body diagonal and six mirror planes, each containing a face diagonal [57]. The F label corresponds to cubic system FCC, following by the corresponds operations.

The symmetry context before exposed can view as a macroscopic symmetry about a crystal system, which means, is very interactive to think as a pattern well-ordered can conform a plane lattice and is intuitively work the symmetry operations. But it's not the only symmetry concept in crystal systems, if we enter into crystal it found atoms or molecules which conforms it. So, the internal study of a crystal add two symmetries to the actual worked before. These "microscopic symmetry" [57] the make reference to \mathbf{k} -space or reciprocal space. So, the previous concept of lattice it's also known as direct space lattice. Thanks to X-Ray, Electron or Neutron diffraction techniques, it was possible to study the internal structure o crystal symmetries in the reciprocal space, this trough diffraction phenomena, the propagation of waves into crystal can to form well defined pattern they which are explained by the wave-vector concept [51, 59]. Therefore, is expected that the electron wave function can be denoted with a lattice periodic part $u(\mathbf{r})$ and wavelike part $e^{i\mathbf{k}\cdot\mathbf{r}}$ so, the set of all wave vectors \mathbf{k} corresponds to plane waves due the lattice, this is known as reciprocal lattice [60]. Then taken into account this, and the vectors \mathbf{a} , \mathbf{b} and \mathbf{c} in reciprocal space can describe the total unit cell [51, 60].

Finally, as a result to get the unit cell in reciprocal space and known which this is composed by lattices, these lattices are called as *Brillouin zones*. Practically the *Brillouin zones* are constructed by drawing the vectors \mathbf{K} defining the reciprocal lattice and then bisecting each of these with planes perpendicular to \mathbf{K} [51]*. In Figure 2.1(b) it's schematized the *Brillouin zone* to GaAs crystal structure, specifically this representation is called as *first Brillouin zone*. To GaAs crystal it was defined the symmetry operations which compose the symmetry elements in Hermann-Maguin notation as the point group $F\bar{4}3m$, it's important to consider the Schoenflies notation also, this due people often speak in terms of both, although the Hermann-Maguin notation is consider as the International notation. In Schoenflies notation, the GaAs correspond to T_d point group.

*The wave vector \mathbf{K} is defined in [61] equation (1.5)

2.1.1 The symmetry and the Band Structure

Returning to Figure 2.1(b), the Brillouin zone have labels which they are, importantly, this is because each of these denote a point group symmetry. These points are: Gamma, X, L, W, U, K. In Schoenflies notation these correspond to: $\Gamma \rightarrow O_h$, $X \rightarrow C_{4v}$, $L \rightarrow D_{3d}$, $W \rightarrow C_{4v}$, $U \rightarrow C_{2v}$, being Γ the high symmetry point. Then, why is the importance of the BZ role in semiconductor band structure?, the answer is the aim of this subsection. We started with the first section of this work, in it refer the importance of solution of Schrödinger equation, specifically at crystal structures as semiconductors. Here, the most important tool is the Bloch's theorem, it which is developed from periodic property of crystal so, it's possible to approximate.

This context is introductory and general, because this doesn't possible if not consider the symmetry properties in crystals, in fact, *the symmetry of system define the basis function to get the electron band structure* [33, 62–64]. Remember that the concept of *basis function* is a mathematical concept, which in quantum mechanics it's known as *Wave functions*. Also, never to forget which the *symmetry* concept is inherent in physics. Therefore, the Group theory establishes the game rules. So, the BZ is the result of Group theory applied in crystal structures, then the BZ is the map to understand the electron behavior in crystal structures, this defined the \mathbf{k} points trough high symmetry paths, where this starts at Γ point or $\mathbf{k} = 0$. If observe the Section 1.1.1 the horizontal axis correspond to \mathbf{k} points and labeled the high-symmetry directions from Γ , then this is the \mathbf{k} paths in BZ as can see in Figure 2.1(b).

Previously, it's continuously mentioned which band structure calculations are difficult, so, it will start to change the hard word to tricky, this because it's possible to make very good models and approximations taking into account the symmetry of the system. All to begins from symmetry, the well-known models to calculate band structure starts from symmetry arguments of crystal or the semiconductor studied, through succession of symmetry operations it knows until it's invariant, this mean doesn't change under transformation. Here, highlights the invariants concept, which is the connection of symmetry and Quantum Mechanics. The symmetry gets the information of the system, while the QM the information of the state electrons. The Hamiltonian of the crystal has a symmetry which depends on their potential, then the crystal potential posses a point group, which is invariant under any transformation. So, the solution of the Schrödinger equation contains the state of the system. From these tools, can propound the physics of electrons or another quasi-particle inside a semiconductor, for example in perturbation theory, starts from Hamiltonian \mathcal{H}_0 with it specific space group, but under perturbation the Hamiltonian of the system should be the sum of $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}'$, where this last has the symmetry correspond to a subgroup of the \mathcal{H}_0 group. This is, the principle of this work which after will be discussed with detail. While the solution of Schrödinger equation with the total Hamiltonian \mathcal{H} will result in the energy spectrum $E(\mathbf{k})$ along of the BZ.

Being a crystal system and the potential is the periodic, it's to hope which a multiband spectrum. Although here doesn't consider the degeneracy* term, it's evident which the Group Theory has the solution, in general words, are the irreducible representations of the symmetry group which determine the dimension of degeneracy [65].

Thus the band structure as a whole exhibits the symmetry characterized by the crystal [65]. All previous it's about of an ideal crystal, then it's possible to get exact solutions of Schrödinger equation. But, to determine in detail the spectrum $E(\mathbf{k})$ throughout the BZ, one needs a numerical solution of the Schrödinger equation. In previous sections, it shows the results of apply TB method to GaAs bulk, this method parts from Bond Orbital Model [36, 66, 67], in this case, the basis functions it's forming linear combinations of atomic orbitals (LCAO) to specific symmetry group [62].

In this method, the importance is the arrangement of atoms and their orbitals considered, for Figure 1.2 these are sp^3 . In another way, in the case of $\mathbf{k} \cdot \mathbf{p}$ method, apart from perturbed model, but in both the main idea it's found the \mathbf{k} -points correspond to the symmetry of the system. In another way, in the case of $\mathbf{k} \cdot \mathbf{p}$ method, apart from disturbance model, but in both the main idea it's found the \mathbf{k} -points correspond to the symmetry of the system. The difference apart from their basis is the efficiency in their applied over semiconductor structures, this means that the $\mathbf{k} \cdot \mathbf{p}$ -method is appropriate in a small region of BZ to describe $E(\mathbf{k})$, therefor is the preferred option to describe semiconductor bands around of Γ , while if the idea is describing $E(\mathbf{k})$ in an extended region of the BZ the TB-method is the correct [62, 65]. In any way, the symmetry establishes the basis to get semiconductor band structure, no matter the method this, includes the first principle methods as DFT, which requires the symmetry information of the system to get the pseudopotentials and the geometric optimization to enhance calculations.

Although the symmetry concept is the base, the principal objective in this work you have to see the consequences of symmetry under in a non-direct perturbation, this mean, if it has GaAs bulk-semiconductor this has one defined point group T_d . When applied it a perturbation, as an electromagnetic perturbation the principal symmetry group it's reducing to a subgroup of itself. This is to mean which the subgroup is also invariant. This conception is also knew or called *symmetry breaking*. It this concept, it was employed at first time by Pierre Curie at ended of nineteenth century [68, 69], It this concept, it was employed at first time by Pierre Curie at ended of nineteenth century, he establishes that, if happens thing which doesn't allow system invariance, so the original symmetry it's lowered then, this mean which symmetry is broken. But, what is the importance to focussing on that?. The importance of symmetry breaking is the physical effects that are presents, the properties of semiconductors changes under reduce symmetry and this can observe in band structure. In the next section will be discussed the importance of symmetry breaking in semiconductors and above all over QWs.

*This is due to the linear independent solutions, which corresponds to one energy, this mean m -fold band degeneracy at the point \mathbf{k} [65]

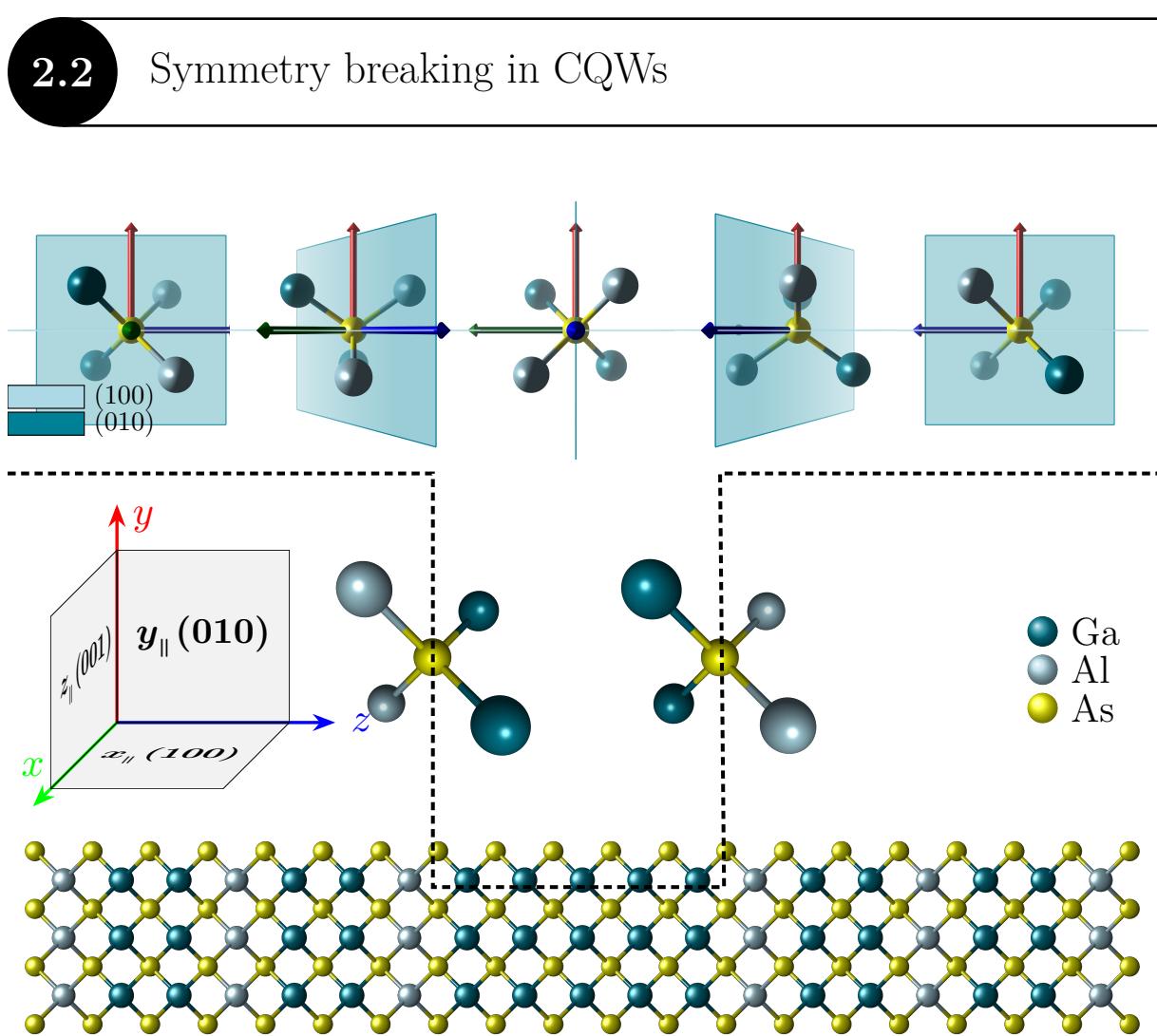


Figure 2.2: In this figure, it's schematized the atom arrangement of $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ single QW. At bottom can see the atom structure assuming the Al concentration to barriers, then, in middle of figure draws the conduction band edge as a potential profile, at interfaces between the change from barrier to well and vice versa, are located the matched atoms, here it can observe that at these positions (interface) the symmetry of changed. At top, it's added the atoms' basis where it has taken the Arsenic as central atom, and scheme the continues rotation over the y -axis.

The symmetry breaking, it's the basis for the physical model in this work. Starting from the general and brief concept of the symmetry importance in the Solid State viewed in the past section will arise the symmetry role and the reduced symmetry in CQWs. Before starting the history, it's important to clarify that the SOC isn't considered in this work, although in the future works of the LFLM are considered, and they study spin phenomena in CQWs. Then, to start in terms of symmetry the GaAs bulk has T_d point group, without intention to minimize the operations only focussing on their subgroups this due as before mentioned if the symmetry is lowered then the point group reduces to one of subgroup [62].

So, in the superlattice case the symmetry reduces at interfaces due to the change of atom species, that is to say, if it parts from GaAs and added an AlAs or $\text{Al}_x\text{Ga}_{1-x}\text{As}$ lattice the new atom structure reduce symmetry elements that can be done then the point group T_d it's reduce. Let's discuss first the simple QW GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ structure grown over [001] direction, if suppose that the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ /GaAs and GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ interfaces* are “perfect”, this mean the QWs structurally are perfect, the symmetry of system it's reduced to from $T_d \rightarrow D_{2d}$ [70, 71].

If taken into account a common atom as in this case the As atom as can see in Figure 2.2 and consider that structures growth (001)-oriented lack of translational symmetry [71], then in a single QW, the translational invariance along z axis is lost [72] as can see in Section 2.2. Is so fact which the visualization of symmetry operations isn't trivial, it can support each other with open software library as Spglib or ASE symmetry functions, these are great tool which has an iterative algoritm under applied recursive operations. If set the arrangement of atoms as the bottom of Figure 2.2, sorting the atoms positions as As-Al-Ga-Ga-As-Al super lattice then at the center of heterostructure the GaAs atoms to finally complement with the heterostructure composed by the first super lattice. Then, it's applies over these the continuous operations under ASE package the result is the $P\bar{4}m2$ symmetry in terms of international notation, therefore the D_{2d} point symmetry group. Although that is well-known and described at [70–76], the use of package software will be got a powerful tool to developed future works. So, in order to understand reducing symmetry from $T_d \rightarrow D_{2D}$ of QW grown on the (001), the $z_{\parallel}(001)$ direction becomes inequivalent to both $x_{\parallel}(100)$ and $y_{\parallel}(010)$ directions, hence the symmetry is reducing.

In therm of band structure the symmetry breaking, or symmetry reducing, generate changes, above all over the VB of QWs. This was expected, the reason is the due to the BZ zone is reduce to Γ point. From here starts the relevance of Γ and the events which occurs are the next: the first one event which occurs is over VB, due to the BZ is reduces to Γ the VB it splits for heavy- and light-holes. This is, from bulk as show the band structure in Section 1.1.1 the VB is four-fold degenerate, then in QWs the heavy- and light- holes bands splits, so, it gets two-fold degeneracy as shows in Figure 1.9. In fact, when refer to Γ in really it's refer to near band extrema, in case of GaAs and $\text{Al}_x\text{Ga}_{1-x}\text{As}$ being direct band gap semiconductors this really clearly. The consequences of bands split it's the reazon which the effective mass approximation (EFA) can works, thanks to that the Schrödinger 's equation can solve as one dimensional equation over both CB and VB band under structure material parameters. Then, the facility of solve Schrödinger 's equation it's limited to only getting information about transitions, it is important to remember which this solution is over real-space, for this reason, it says that the potential profile is called a band edge profile and can't confuse with the band structure. So, this is the principle of symmetry reduction in QWs structures, in the next section will be addressed the symmetry in CQWs structures and the mechanisms to reduce it.

*Another nomenclature usually used is A/B interfaces, being reference to two dissimilar atoms.

2.2.1 Coupled Quantum Wells

The symmetry breaking doesn't only wight in mathematical aspect, for physics this concept with leads to discover many important and exotic phenomena. When a system reduces its symmetry, their physical properties changes. It's the change in physical properties which reason to study it, in semiconductors as QWs the symmetry breaking starts with modify their band structure (fingerprint) splitting the valence band and reducing the degeneracy, although it can be the first one to appear their consequences get more relevance.

In therms to study changes in physical properties in this work it's focus on optical properties, the light matter interaction gets information about the symmetry breaking through transitions. The GaAs QWs as a direct band gap semiconductor are excellent platform to study light-matter interaction and the effects on symmetry. It's important to remark that from here the excitons played it an important role, in fact, over them falls the physical interpretation of optical properties.

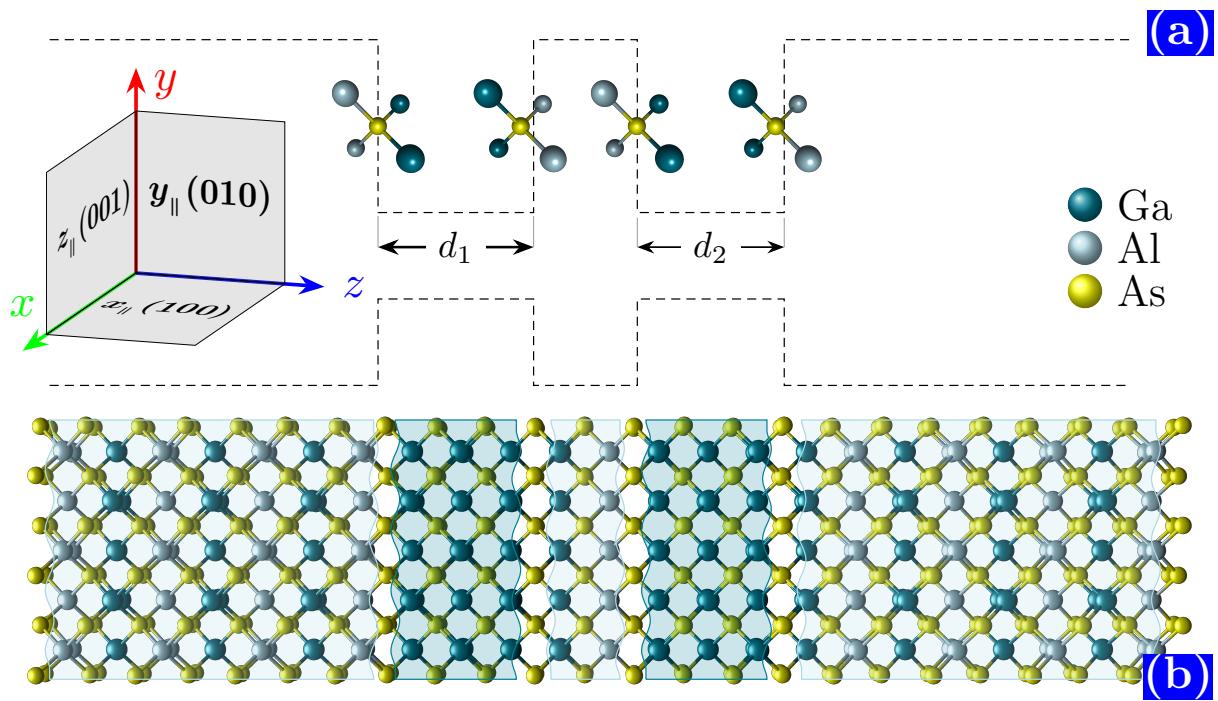


Figure 2.3: General scheme to describe a SCQWs structure. In this case, the barriers are it composed by $\text{Al}_x\text{Ga}_{1-x}\text{As}$ and the wells are of GaAs with the same width ($d_1 = d_2$), while the coupling barrier also composed by $\text{Al}_x\text{Ga}_{1-x}\text{As}$. In top (a) denotes both CB and VB edges profiles over z -axis (Real-space) direction. Then in bottom (b), it represents the atoms structure to CQWs.

Then, in the case, of Coupled Quantum Wells (CQWs) through excitons get optical properties really it's interesting, although, to get that properties the CQWs should get a double reduce symmetry from T_d , even if that's not obtained it's important to anisotropy

spectroscopy get it as a basis. Then, firstly, it's start with the symmetric coupled quantum wells (SCQWs), these are QWs with same width and coupled with a thin barrier, this barrier must be enough thinner so that electron wave function can be overlapping along potential of the two wells. For these structures the symmetry is also D_{2d} as in single QW, it's important to say which, if they aren't consider the interface defects as roughness it's possible to consider ideal interfaces, then the same symmetry operations works in both single QW like a SCQWs.

From Figure 2.3 it's possible to compare which in contrast with the SQW (Figure 1.8) is the same case with exception to the two QWs, although it's true that the technology of growth it's really accurate, the interfaces aren't be exempt free of them. Even, the rough by possible of Al impurity can cause the possible segregation of this, then it's important the Al concentration x [77, 78]. Even tought, an interface grown over (001) is C_{2v} , if consider structurally perfect as SQW or SCQWs, the overall symmetry of both interfaces is D_{2d} [70].

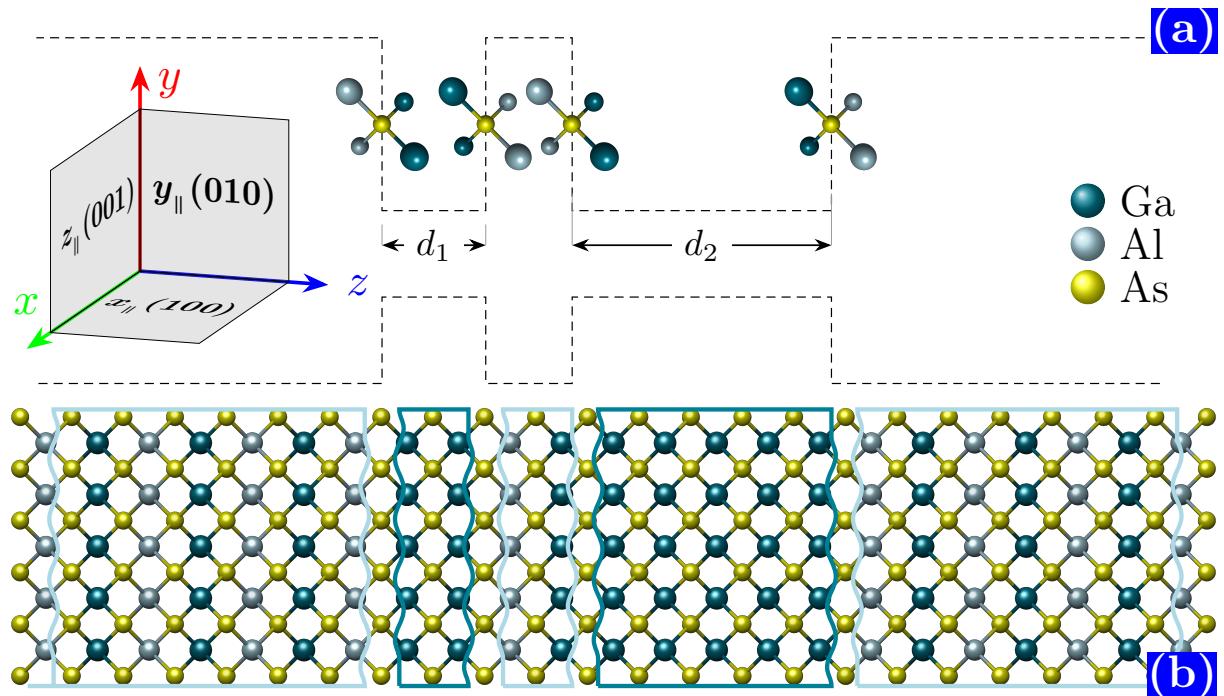


Figure 2.4: General scheme corresponds to ACQWs structure. As in Figure 2.3(b), the structure is basically same both in barriers composition and dimensions as well as the coupling barrier, only changes the relative position of this. Then, result that one of QW is wider than other ($d_1 < d_2$), so, this reason causes the asymmetry in structure. Also, here in top (a) draws the potential profile along z , and bottom (b) scheme the atoms structure where is clear that only changes coupling barrier relative position with respect to Figure 2.3(b)

Remember that as Courie mentioned [68, 69, 79]: a system under perturbation reduces their symmetry to a subgroup of original symmetry group, then, if now starts with D_2 symmetry

this subgroup can only reduce to a subgroup of six possibles, into them is C_{2v} subgroup. Previously it mentioned which, exist several mechanisms can reduce the symmetry, these are usually called perturbations. These perturbations can be nature by different sources, *in this work has been it found a novel source which reduces the symmetry, in other words, broken symmetry without needed external source as applied electric or magnetic fields.*

In the next section, it details the reason which it's called a novel source of reduce symmetry, therefore, before continue it's important to mentioned that the simple reason of modify the one well width in CQWs structures makes the system loses fourfold rotations over $z_{\parallel}(001)$ then, the symmetry it reduces.

If compares Figures 2.3 and 2.4 it's clearly that the representative part of the coupling barrier only shift over z , this allows to simulate a ACQWs heterostructure, this mean which QW is wider than the other, so, it gets an asymmetric structure which along z losses the rotation symmetry. Also, if it's uses symmetry code packages as ASE or Spglib which applies consecutively symmetry operations to both CQWs structures, it results in a D_{2d} and C_{2v} for SCQWs and ACQWs respectively.

2.2.2 Special symmetry reduction from $D_{2d} \rightarrow C_{2v}$

The importance of C_{2v} point group around of QWs system is attractively to study properties of them, over all quantum properties as “spin” [13, 73, 76, 80–82]. Also, it's very important the quantum mixing which exhibits as a result of symmetry breaking, in fact, latter in Section 3.2.3 it presents the result of RAS experiments, which are the result of hole mixing. The anisotropy experimental measured it's caused due to the mixing at VB, therefore it's a direct cause of symmetry. Figure 2.5 shows and schematics the roadmap to get a QWs structures with C_{2v} symmetry. This starts at the left side with asymmetrical structures Figures 2.5(a) and 2.5(b), this asymmetry is related with the potential, exists a variety of them but the objective it's practically the same, the asymmetric potential profile it can be obtained by: asymmetric barrier, this can be due to the change of semiconductor type between adjacent barriers, i.e., AlAs/GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ structure, this can interpret as high barrier/well/low barrier [12]. Another way of them, is caused of a barrier it's under gradient composition [14, 15]. The end case under asymmetric potential consideration shows in Figure 2.5(c), this case is due in one of the barriers is intentionally “inserted” an atom from other specie [16], this causes an asymmetric potential profile. Finally, it display the our CQWs structure, in comparison with all above firstly it has two wells, therefore reduced symmetry it be

Continued with the map, they have the perturbed structures, they are called like that due to they are under an external perturbation, at center top Figure 2.5(d) it's outlined a QW structure under strain applied, while at bottom center Figure 2.5(e) it's showing a QW structure under electric field applied. So that, in both exists, an external perturbation

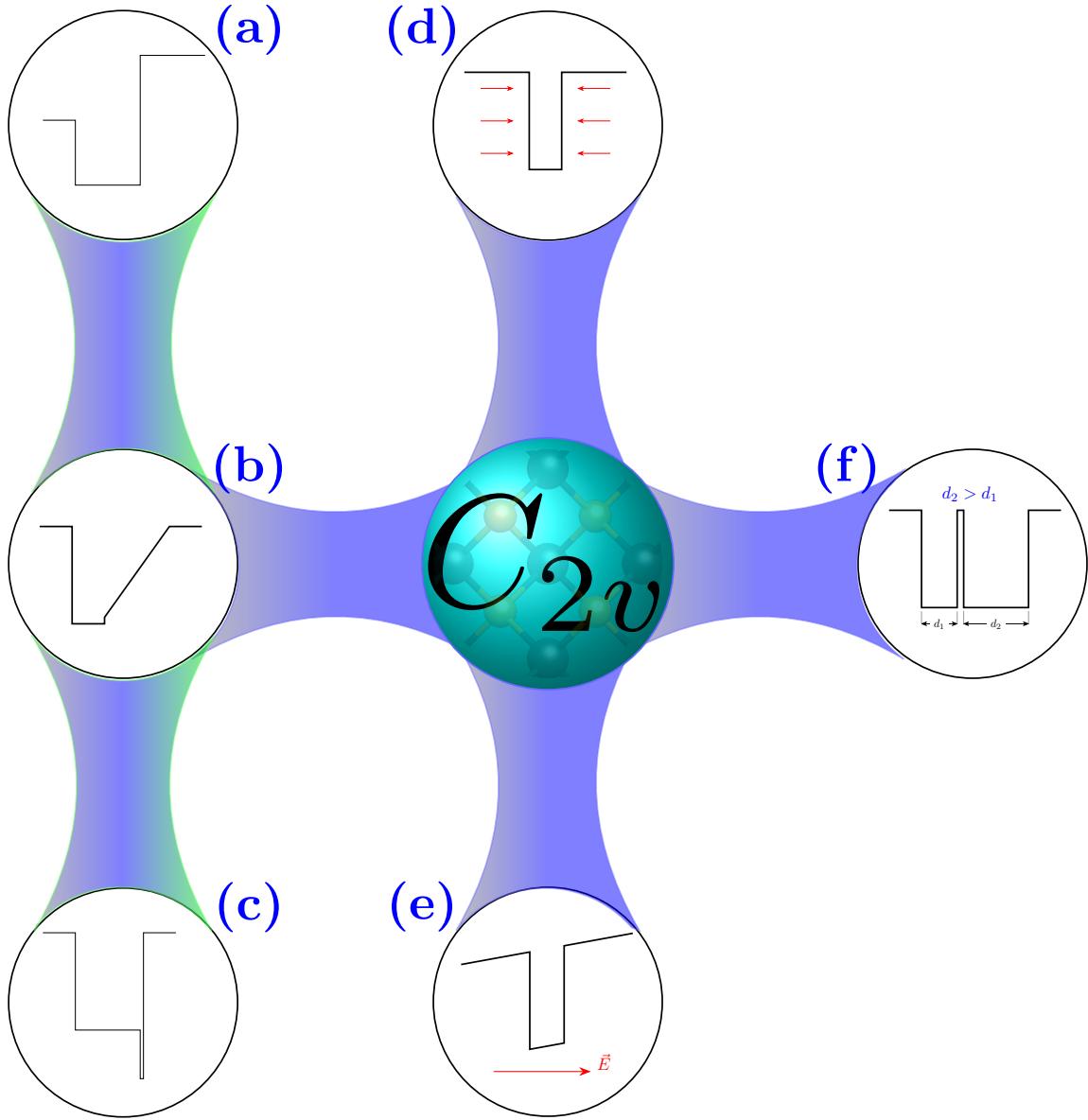


Figure 2.5: This roadmap it's developed around of QWs with remarkable potential profile, this mean posses a desire C_{2v} symmetry. It starts with asymmetric QW (a) [12, 13] specifically in potential, this is due to the semiconductor difference in adjacent barriers to well. At center left (b) and bottom left(c) are asymmetric QWs, in the first case the asymmetry [14, 15] it's caused by gradied concentration in a barrier and bottom while the other case the asymmetry it's originated by insert an atom specie different of the barrier [16]. At top center (d), it's show the QW under strain [17–19] which causes the reducing symmetry, while at bottom center(e) it shows the CQWs under applying electric field [20], the relation of both, it's the external perturbation which causes the breaking symmetry. Finally, at right (f) it's presented the ACQWs which has a symmetry breaking due to the relative width of QWs [21].

which causes a losses symmetry. Finally, it displays the ACQWs structureFigure 2.5(f), in comparison with all above. It's notably that this structures has two wells which are coupled by a thin barrier, by this reason as called Coupled Quantum Wells. Then, which is the reason by these structures are novels?.

To discuss this answer, it's important to mention the relevance of CQWs being that these structures are recurrent studied to observe quantum phenomena as exciton (**X**) condensation [83–85]. It's to be expected that in CQWs can measure indirect transitions, this means that in comparison with a single QWs whereby exists direct transitions (band to band) hardly can measure these. But, the reality of the importance of CQWs being that excitons are very interested to apply in semiconductor devices, the properties of excitons and their interactions really exhibit quantum attractive properties. So, unlike with single QW in CQWs the life of excitons increases [86, 87], in fact, this is one of the principal reasons which that are attractive structures [83, 88, 89]. Also, in terms of spin properties, the CQWs exhibit great potential [90]. Therefore, it being can discuss several interesting quantum properties in comparison that single structures. But, in comparison between CQWs, the symmetric structures need it perturbed to them exhibit these properties, while asymmetric structures are an excellent platform to study quantum properties such as holes mixing, spin, etc. It's then *ACQWs an interesting structure which part from being artificial, to get natural perturbation**, even though, as can see doesn't are the unique structures with “natural perturbation” which generates a symmetry breaking, all above mentioned it's reduced to confinement way. This, is the reason to call special symmetry reduction in a structure hardly studied in several sub-areas of solid state.

2.3 Numerical Calculations

All the above cool properties mentioned, could not have been predicted or observed without their knowing their electronic properties. This is many times mentioned, is the fingerprint of semiconductors, so, also mentioned the problem to calculate it. Here was implemented a *simple* model-based in EFA method to calculate the confined energies in CQWs structures. Before explaining the numerical method to obtain these solutions, it is important to discuss the reason which can apply this method, so, it starts with the definition of band-edge from energy dispersion. In Section 1.1.1 are discussed in general the oncept of VB and CB, also is mentioned the significant methods to calculate them, in case of a heterostructure as CQWs the complexity is greater than bulk case. In bulk, the case is well-known several methods to calculate bandstructure, where all of them are developed by the symmetry properties of the system at stake. In the case of the heterostructure, the symmetry is also important, the problem is developed a Hamiltonian capable to describe all of the

*Thanks Dr. Raul for magnificent description.

system, this means, building a Hamiltonian which considers all properties as symmetry, perturbations, and in this case the potential. This work is very hard, even though exists general Hamiltonians to heterostructures, these don't warranty the correct solutions. The history and development of methods and techniques to calculate electronic properties is an area in constant evolution, from the fifties with Kane [22], Luttinger and Kohn [91] in perturbed methods as $\mathbf{k} \cdot \mathbf{p}$ to Slater and Koster [67] which proposed an atomistic method based on a linear combination of atomic orbitals called as TB, all of this already discussed and mentioned. So, we developed a model to calculate the confined energies in coupled structures. Even though exist analytical methods to calculate confined energies, in case of CQWs is more difficult to get an exact analytical method, while also exist some analytical methods under approximation description [92–94]. In the Section 1.2.2 it's developed the analytical solution of a simple quantum well taken into account the EFA method. It's possible to use this type of methods due to the symmetry reduction from bulk to QW, this due to the split in VB which passes from being a fourfold degenerate to be a twofold degenerate. It's precisely in VB where is it complicated to solved it.

The Figure 2.6 it's the result of apply $\mathbf{k} \cdot \mathbf{p}$ method, specifically taken into account 8-band model Hamiltonian [22, 35, 95] which is an extension of four band model [95] and even, this model is raised to bulk semiconductors, but in Figure 2.6(b) shows the results applied in a GaAs SQW heterostructure. To give support to these calculations is important to invoke EMA, this method is an efficient method to computational calculations [39, 96] and their basis allows developed electron solutions. In general, exist a variety of Hamiltonian intending to calculate the bandstructure of heterostructures. From conventional Luttinger-Kohn model, [91] to relatively recent models by Burt and Foreman [97–99], which they consider the basis functions depends on symmetry, this is the reason which Hamiltonian is associated to the bulk structure as in this case is Zinc-Blende type.

Although it has been discussed applying this method over heterostructures as the QWs, this work considers it as an “exploratory” tool. The reason for taking him into account in that way is basically their difficult and tedious way to get a correct Hamiltonian that describes our structures, in fact, $\mathbf{k} \cdot \mathbf{p}$ isn't the unique difficult method, also TB and others, have laboriousness way to build their Hamiltonians. So, we take the technic and equations from Vurgaftman [35] and solve it numerically to gets the GaAs bulk as shown in Figure 2.6(a) and in the same route, we take the Hamiltonian from Marchewka [23, 100] and solve under FDM, without major preamble the idea is to denote the split between heavy- and light-hole bands which is the justification to our numerical calculations proposed. In Equation (2.1) it's presents the 8×8 Hamiltonian solved to get bandstrcuture to GaAs SQW.

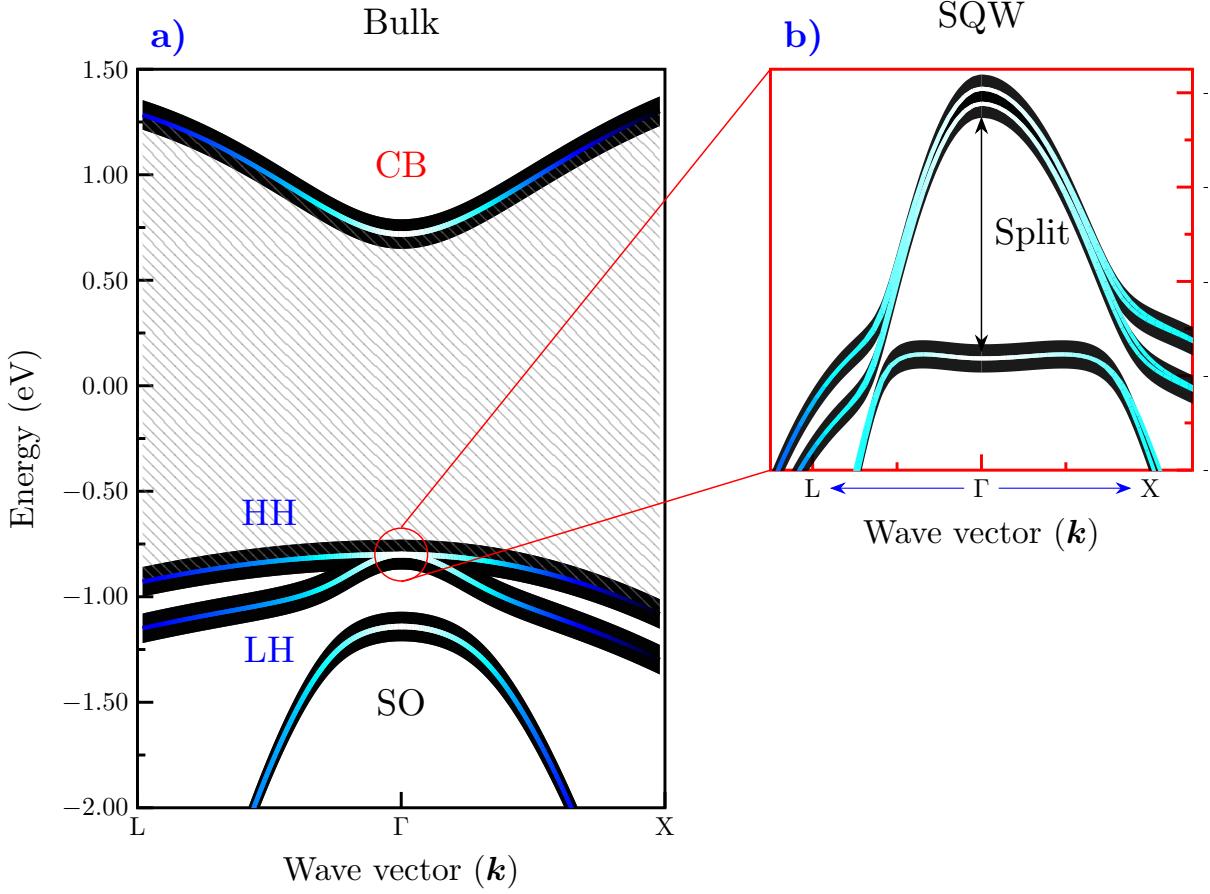


Figure 2.6: (a) The GaAs band structure calculated with 8-band Khane Hamiltonian [22, 23], therefore SOC is consider. At $\Gamma = 0$ in Bulk VB is closed by a circle, here observed the degeneracy, while in (b) it's denoted the split (Δ) between heavy- and light hole bands. Also due SOC consideration, it displays the spin up and down, bands.

$$H_0 = \begin{bmatrix} T & 0 & -\frac{1}{\sqrt{2}}Pk_+ & \sqrt{\frac{2}{3}}Pk_z & \frac{1}{\sqrt{6}}Pk_- & 0 & -\frac{1}{\sqrt{3}}Pk_z & -\frac{1}{\sqrt{3}}Pk_- \\ 0 & T & 0 & -\frac{1}{\sqrt{6}}Pk_+ & \sqrt{\frac{2}{3}}Pk_z & \frac{1}{\sqrt{2}}Pk_- & -\frac{1}{\sqrt{3}}Pk_+ & \frac{1}{\sqrt{3}}Pk_z \\ -\frac{1}{\sqrt{2}}k_-P & 0 & U + V & -\bar{S}_- & R & 0 & \frac{1}{\sqrt{2}}\bar{S}_- & -\sqrt{2}R \\ \sqrt{\frac{2}{3}}k_zP & -\frac{1}{\sqrt{6}}k_-P & -\bar{S}_-^\dagger & U - V & C & R & \sqrt{2}V & -\sqrt{\frac{3}{2}}\tilde{S}_- \\ \frac{1}{\sqrt{6}}k_+P & \sqrt{\frac{2}{3}}k_zP & R^\dagger & C^\dagger & U - V & \bar{S}_+^\dagger & -\sqrt{\frac{3}{2}}\bar{S}_+ & -\sqrt{2}V \\ 0 & \frac{1}{\sqrt{2}}k_+P & 0 & R^\dagger & \bar{S}_+ & U + V & \sqrt{2}R^\dagger & \frac{1}{\sqrt{2}}\bar{S}_+ \\ -\frac{1}{\sqrt{3}}k_zP & -\frac{1}{\sqrt{3}}k_-P & \frac{1}{\sqrt{2}}\bar{S}_-^\dagger & \sqrt{2}V & -\sqrt{\frac{3}{2}}\bar{S}_+^\dagger & \sqrt{2}R & U - \Delta & C \\ -\frac{1}{\sqrt{3}}k_+P & \frac{1}{\sqrt{3}}k_zP & -\sqrt{2}R^\dagger & -\sqrt{\frac{3}{2}}\bar{S}_-^\dagger & -\sqrt{2}V & \frac{1}{\sqrt{2}}\bar{S}_+^\dagger & C^\dagger & U - \Delta \end{bmatrix} \quad (2.1)$$

where

$$\begin{aligned}
k_{\parallel}^2 &= k_x^2 + k_y^2, \quad k_{\pm} = k_x \pm ik_y, \quad k_z = i\partial/\partial z, \\
T &= E_c(z) + \frac{\hbar^2}{2m_0} \left[(2F+1)k_{\parallel}^2 + k_z(2F+1)k_z \right], \\
U &= E_v(z) - \frac{\hbar^2}{2m_0} \left(\gamma_1 k_{\parallel}^2 + k_z \gamma_1 k_z \right), \\
V &= -\frac{\hbar^2}{2m_0} \left(\gamma_2 k_{\parallel}^2 - 2k_z \gamma_2 k_z \right), \\
R &= -\frac{\hbar^2}{2m_0} \sqrt{3} \left(\mu k_+^2 - \bar{\gamma} k_-^2, \right), \\
\bar{S}_{\pm} &= -\frac{\hbar^2}{2m_0} \sqrt{3} k_{\pm} \left(\{\gamma_3, k_z\} + [\kappa, k_z] \right), \\
\tilde{S}_{\pm} &= -\frac{\hbar^2}{2m_0} \sqrt{3} k_{\pm} \left(\{\gamma_3, k_z\}, \frac{1}{3} [\kappa, k_z] \right), \\
C &= \frac{\hbar^2}{2m_0} k_- [\kappa, k_z]
\end{aligned}$$

and each of these parameters taken from [35]. The Hamiltonian H_0 is defined for [001], and the principal idea is evaluating it in a “discrete” SQW structure, the ”discrete“ term refers to a numerical technique for solving it. The results it shows in fig. 2.6(b), this approximation is enough to evince the VB split, even, the results also CB information the importance falls on VB, this is due that difficult to solve there. Then, the VB split allows applying “simple” numerical methods without needing to define complex Hamiltonian and many parameters. In next section it will discuss the physical and mathematical formulation to calculate the electron energy confined, beginning with the symmetry reduce causes the VB split and developed all around of $\Gamma = 0$ this means we work over band-edge potential in real space.

2.3.1 Envelope Function and Effective Mass Aproximation Methods

Furthermore, to get a numerical method and solution robustness without any complex formalism but based on the essence of this work which is the effect of asymmetric width potential in CQWs which causes the symmetry breaking and this physical property involve important optical phenomena, as increase Anisotropy depending on relative width in these. The EFA is the mathematical justification to model electron behavior under periodic potential as a crystal, while the EMA is the physical model and the considerations to study the electron behavior inside a periodic potential from a crystal [39]. Under that

*Frequently the EFA and EMA approximations are considered as the same term, although maybe being correct, given that one depends on the another, is important to remark that the EMA depends on the formalism of EFA.

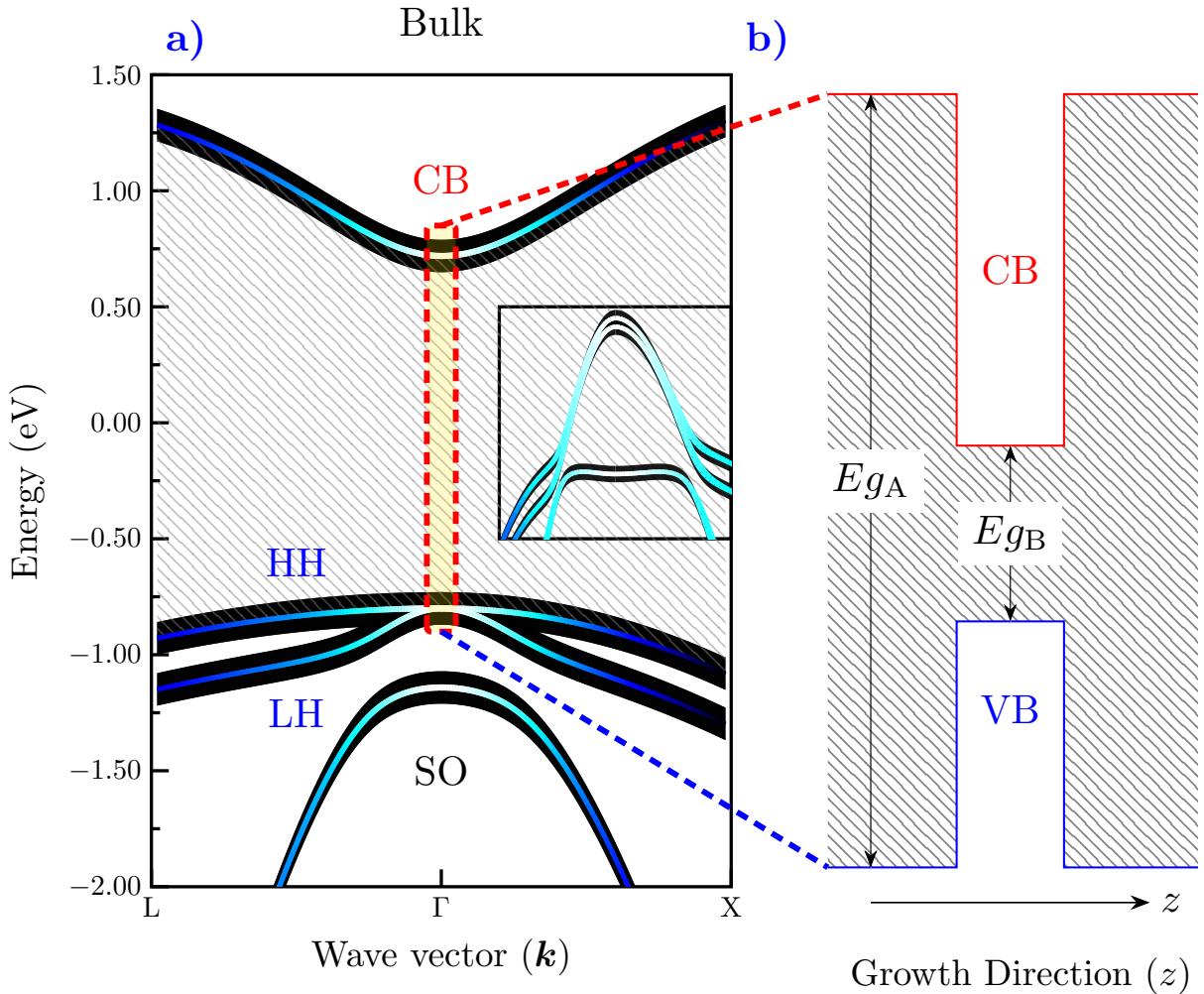


Figure 2.7: (a) it's tyhe focus over $\Gamma = 0$ and inside is the SQW $\mathbf{V}\mathbf{B} \cdot \mathbf{p}$ results, while the

assumptions, it's possible to review several considerations, that considerations reduce the complexity of solutions and allow simplifying the calculations. Then, in all of this work it develops around of $\Gamma = 0$, this means that will use a one-band model. To resume this last context, it's importantly to taken into account the next considerations. Firstly, from here we can study the electron and holes behavior by separately, from basic assumption which $\Gamma = 0$ this enables to pass from electronic band to band edge (see Figure 2.7). So, it's important to define that tools previosuly mentioned. The EMA is a valid approximation in bulk materials, in fact this is an elemental model which due to the good results and simplicity it's can be applied over heterostructures. About of this last, the heterostructures are complex, the dissimilar matched semiconductors which conformed that it, posses doesn't only difference in band-gap energy but also the effective mass.

If join all of that parameters involved it, the model to solve should being proportional to the considers in it, by this reason models as TB are complex due to the difficult to define

all that parameters, while in $\mathbf{k} \cdot \mathbf{p}$ are well-defined, but contrary, the method frequently is submitted to discussion due to the assumptions as the doesn't "see" the correct point-group symmetries, in fact, frequently assumes which the system already owns it. We can introduce ourselves in a severe discussion about of bandstructure calculations methods, but the reality is can't define a standard method, so that, doing reference to Harrison et al. [39], we can apply the simplicity method. Then, assuming the simplicity term, in which practically mentioned that, it's important to consider the simple way to get a result as a long as this works. So, in this work, will discuss the increase of the IOA due to the relative width in the wells of the coupled system, then here only considers the potential profile as a really significant parameter, of course, the effective mass and the band-gap are also essential, but these are considered as a part of the model. Being this the basis of the model, this work will employ the formalism of EFA where in a general explain, this it gives the envelope functions to calculate the single electron behavior under periodic potential as a heterostructure, therefore, this can allow describing it through position-depend material properties [43, 101, 102], then it's also indispensable to declare the boundary conditions owned by the EFA. Therefore with a mathematical formalism, then it's employed the EMA which parts of the fundamental parameter which is the effective-mass, then it can work with the 1D-Schrödinger 's equation as a function of effective-mass, as traditional eigen-value equation:

$$\mathbf{H}\psi(z) = E\psi(z) \quad (2.2)$$

Then, the Hamiltonian of EMA it's developed mainly by the effective-mass, this has the advantage of evaluated along of heterostructure, allowing the change of semiconductors which composed that, so, this makes it a great method. Therefore the Hamiltonian for a particle is given by [43, 101, 103]:

$$H_z = \frac{p_z^2}{2m^*(z)} + V(z). \quad (2.3)$$

In Equation (2.3) it's taken z -axis as a direction perpendicular to interface of QWs, or simply as the growth direction, consequently the potential $V(z)$ it's depend on, and the effective-mass $m^*(z)$ also depends on position z . The Equation (2.3) it's taken z -axis as a direction perpendicular to interface of QWs, or simply as the growth direction, consequently the potential $V(z)$ it's depend on, and the effective-mass $m^*(z)$ also depends on position z . As can see in Figure 2.7(b), it's showing the band-edge profile as a potential profile which depends on semiconductor band-gap along of heterostructure. Also in Equation (2.3), it considers the momentum as p_z^2 as a 1D, due to the before discussed. Then it's allow to developed a method to solution which depends of position z . Finally, it can denote a 1D Schrödinger quation under EMA to electron also heavy- and light-holes :

$$\left[-\frac{\hbar^2}{2m_{jz}^*} \frac{d^2}{dz^2} + V(z) \right] \psi_{nj}(z) = E_{nj} \psi_{nj}(z), \quad (2.4)$$

$j = e, hh, lh.$

The Equation (2.4) it's then the effective-mass equation implemented in this work. The solution of that, it's detailed in next section where explains the numerical method to solve it. Before to continue, it's important to discuss the 1D equation. This equation will be applied over each particle ($j = e, hh, lh$), this means, over electron, heavy- and light-holes, where only considers the effective-mass (m_{jz}^*) trough each semiconductor in heterostructure studied and the most important in this work, the potential profile $V(z)$ which depends on relative widths of coupled wells.

2.3.2 Finite Difference Method

In this section it will discuss the numerical method to solve the 1D Scrödinger 's equation, even tough this is a large discussed theme, here focus on a simple but powerful numerical method. Started from the fact that the Equation (2.4) can it solve along of heterostructure and taken into account each semiconductor with compose it, this means that can discretize that and solved it for electron and holes. Although this, being a 1D equation, doesn't remove importance in the solution, that is to say, it's needing a robust method. It's important to remark which the spacial discretization depends on δz .

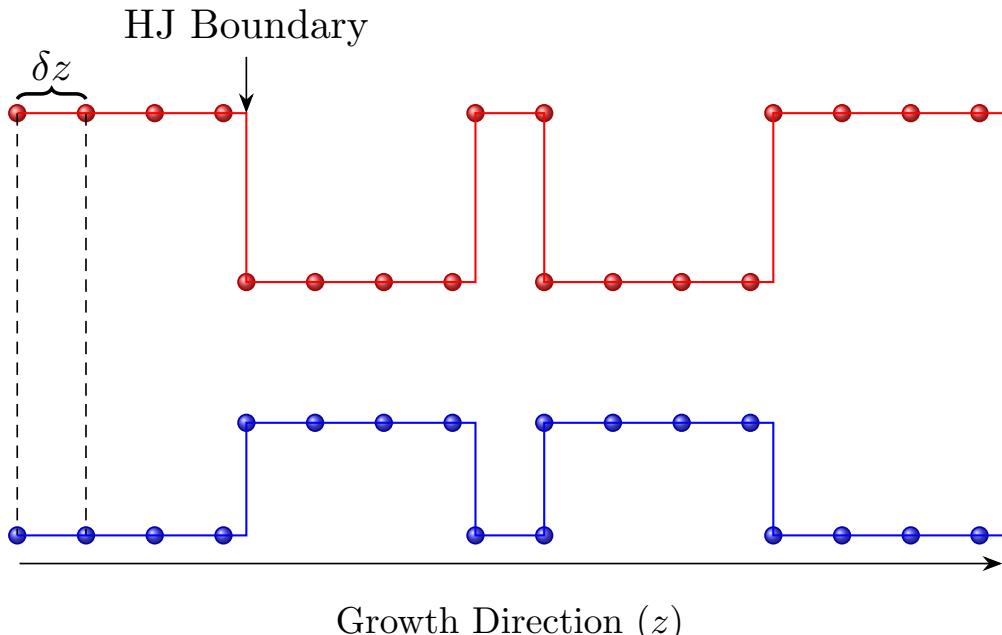


Figure 2.8: Sketch of discrete potential band-edge profile which depends on spacial desplacement δz . HJ denotes the Heterojunction boundary.

In Figure 2.8 schemes the discrete potential profile considers, this band-edge profile corresponds to a SCQWs structure. Remember that the potential $V(z)$ comes from the bend-edge Energy E_{edge} , like in the CB and VB, from that take the effective mass which corresponds to each semiconductor which compounds all heterostructure. Then, we can divide a heterostructure with coupled quantum wells basically into two parts, the adjacent barriers and the zone of wells which is considered the coupled barrier and their width as well as the relative widths of each well. So, if consider the discrete structure we can apply Equation (2.4) in each point which composes that.

The solution of Equation (2.4) it's the central discussion, although exist several numerical methods to solve it, in this work is employed the *Finite Difference Method* (FDM), which begin characterized by the easy computational application. Even if, this method doesn't unique in applied discretization in comparison with the other method as Shooting [39, 104], reduce the computational time and assure the correct convergence as long as the boundary conditions are correct. In other words, the FDM is a non-sensible convergence method, being focused on matrix solution. Taking the Equation (2.4) and reformulated as a *difference equation* [39]:

$$-\frac{\hbar^2}{2m^*} \left[\frac{\psi(z + \delta z) - 2\psi(z) + \psi(z - \delta z)}{(\delta z)^2} \right] + V(z)\psi(z) = E\psi(z). \quad (2.5)$$

In the Equation (2.5) it's denoted the derivative change by numerical difference, being a discrete equation. Something that it's important to consider is the boundary conditions which depend on the number of semiconductors involves in the structure and their dimensions, then, if it requires high precision it's important two factors: the total dimension of the structure, which contemplates each heterojunction boundary and the spatial step defined by δz . Either way, if considering a wide structure, as well as a very small step ($\delta z \approx 1\text{\AA}$) the computational effort, is high, for this reason, the compute considered memory should be reasonable. Then, taking into account these two factors and considering the computational work, the best choose a Matrix solution.

The Matricial solution allows solving the system of equations naturally as an eigenvalue problem, then it has several numerical alternatives* to solve the system. The reason to assert these has to do with the discrete solution as large as the ratio between structure width and spatial step. This means the matrix has dimensions as : $N = \text{total width}/\delta z$. Then, the way to get matrix (\mathbf{H}), from Harrison et al. [39], consists in reformulate Equation (2.5) as function of spatial potential band edge taken into account the variable effective mass, then:

$$a_i\psi_{i-1} + b_i\psi_i + c_i\psi_{i+1} = E\psi_i \quad (2.6)$$

*This refers to linear algebra techniques [39, 105]

where the coefficients are:

$$a_{i+1} = c_i = -\frac{\hbar^2}{2m_{i+\frac{1}{2}}^*(\delta z)^2} \quad \text{and} \quad b_i = \frac{\hbar^2}{2(\delta z)^2} \left(\frac{1}{m_{i+\frac{1}{2}}^*} + \frac{1}{m_{i-\frac{1}{2}}^*} \right) + V_i \quad (2.7)$$

From Equation (2.2), then the matrix \mathbf{H} is conformed by the system of equations which is evaluated in each point os structure, then \mathbf{H} is:

$$\mathbf{H} = \begin{pmatrix} b_1 & c_1 & 0 & \cdots & 0 \\ a_2 & b_2 & c_2 & \cdots & 0 \\ 0 & \ddots & \ddots & \ddots & 0 \\ \vdots & \cdots & a_{N-1} & b_{N-1} & c_{N-1} \\ 0 & \cdots & 0 & a_N & b_N \end{pmatrix} \quad (2.8)$$

then ψ is in Equation (2.2) is a vector column which containing all the samples of the wave function.

Then, for the solution of the $N \times N$ matrix it's represents a computation work, but due to the *tridiagonal* nature it reduces that effort and thanks to exist great algebraic algorithms to solve [106, 107], this work can simplify enormous. Therefore, this method simplifies the solution of any structure with arbitrary or in our case dependent on band-edge potential. The next task is over to define the potential profile and their dependence of effective-mass and band gap energy (E_g). Firstly, we take as reference the double quantum wells propose by Harrison et al. [24] and reproduce the eigenenergies and wave functions as shows in Figure 2.9. For this structure, it takes into account symmetric double wells, that is to say, with same width $l_w = 6\text{nm}$, $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ central barrier of $l_b = 4$. For the semiconductor parameters with compose that structure, as the model like shows in the Equation (2.4) doesn't take into account the effective mass as energy function and not taken into account the nonparabolicity [108, 109]. Then, to define potential profile $V(z)$ uses the Varshni's model [110], to calculate bandgap energy to $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$, how doesn't consider the mass-effective as energy dependence then it uses paramaters from: [111–114], finally it employs the Vegard's law to define the thernary semiconductors [115, 116]. If defines the potential profile as:

$$V(z), m^*(z) = \begin{cases} \text{Al}_{0.2}\text{Ga}_{0.8}\text{As} & 0 < z < 20 \text{ nm} \\ \text{GaAs} & 20 < z < 26 \text{ nm} \\ \text{Al}_{0.2}\text{Ga}_{0.8}\text{As} & 26 < z < 32 \text{ nm} \\ \text{GaAs} & 32 < z < 38 \text{ nm} \\ \text{Al}_{0.2}\text{Ga}_{0.8}\text{As} & 38 < z < 58 \text{ nm} \end{cases} \quad (2.9)$$

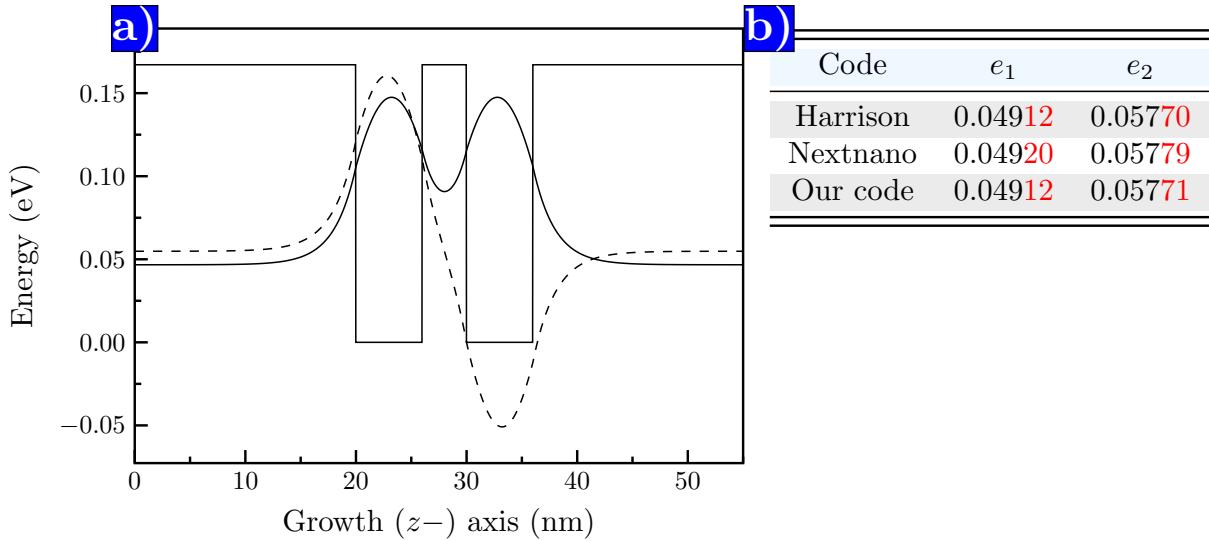


Figure 2.9: Doble Quantum Wells structure reproduce from Harrison et al. [24], (a) shows the plot to each energy calculate e_1 and e_2 , while in (b) the table exposes the comparison of numerical results from **Harrison** et. al., **Nextnano** software [25] and our results. The difference taken into account the Harrison’s results practically are the same, but around of meV orders our results are precisely.

where the $V(z)$ it defines by the correspond value of E_g and the effective-mass $m^*(z)$, both with the spatial z dependence. But, this work as only in take **Harrison** et al. [39] like a reference, the **Nextnano** software [25, 117] also take that, in the Figure 2.9 and Figure 2.9(b) shows the table of comparison results, taken as basis **Harrison** et al. and compare also with **Nextnano** software and the codes developed in this work [1]. As can see, the results getting in this work are precise with respect to Harrison, and it’s important to remark this work doesn’t intend to denote supremacy since that fall short of sense, the intent is only the comparison to demonstrate that our codes works. The table in Figure 2.9(b) it denotes (with red) the precision around of meV, this is what we mean when say the major precision with respect to Harrison’s results. Thus, we can trust the codes and model here present, do they work concerning with computational aspect and the physical results, and although only focused over electrons’ solution in the band-edge profile, the next part of this works presents the results obtained in the interest structures.

2.4

Numerical Results

Here, we introduce the results obtained in CQWs studied in this work. Section 3.1 it’s detailed of the structures’ composition as well as the properties of these. Then, here focus only in the numerical and computational results, over all, in the calculation of confined

energies it remembers that our model is oriented only about energies, so that, frequently explain many parameters and arguments that doesn't consider here. But, that doesn't mean which the obtained results minimize the quality of this work, in fact, we achieve very interest results without need to draw on to the very hard models. The Figure 2.10 shows the plot of wave functions for electrons, heavy- and light-holes resultant of numerical calculations, here taken into account only four samples detailed in Section 3.1 at the Table 3.1, all of these samples are the CQWs structures studied in this work, then over each of these was calculated the confined energies and the wave functions profiles. In the Figure 2.10 it's calculated ψ to denoted more clarify the overlapping wave functions over two coupled wells, even tough here doesn't show the total table of confinement energies results, later these will compare with the experimental transitions. Therefore, in this section, we limited to specify the numerical results in accordance with the published work [21].

Sample	e1-hh1	e1-lh1	e2-hh2	e2-lh2	e3-hh3
SCQWs	(N)1.5328 (E)1.5297	(N)1.5380 (E)1.5341	(N)1.5394	(N)1.5499	(N)1.5948
ACQWs-1	(N)1.5265 (E)1.5273	(N)1.5296 (E)1.5314	(N)1.5368	(N)1.5460	(N)1.5837
ACQWs-2	(N)1.5181 (E)1.5190	(N)1.5206 (E)1.5210	(N)1.5330	(N)1.5394	(N)1.5460

Table 2.1: Direct transitions (**X**) calculated for two ACQWs and one SCQW detailed in Section 3.1 and Table 3.1. From up to down shows the numerical (E) and experimental results, the experimental results was obtained from RAS experiments which are performed at 30K.

In the Table 2.1 exposes and compares the numerical results obtained, it's importantly to remark that the calculations were performed at 30K, this due to the central experiments (RAS) are performed to that temperature, by this reason the parameters involves are well-defined as a function of temperature. These calculations taken into account the conduction and valence-band offsets of 65% and 35% respectively. Both electron and hole effective masses for GaAs and AlAs can be found in Refs. [111–113], whereas the ternary $\text{Al}_x\text{Ga}_{1-x}\text{As}$ we used the Vegard's law [115]. For the numerical direct transitions (**X**) calculated, it's consider the exciton binding energy as a function of well width [118, 119], as commonly only shows the direct transitions in a range of wells widths, we interpolate these energies to used in accordance with the structures used in this work. The Figure 2.10 denotes some interesting physical issues, one of these with principal role in this work it's a linked between barrier width and the relative width of the coupled wells.

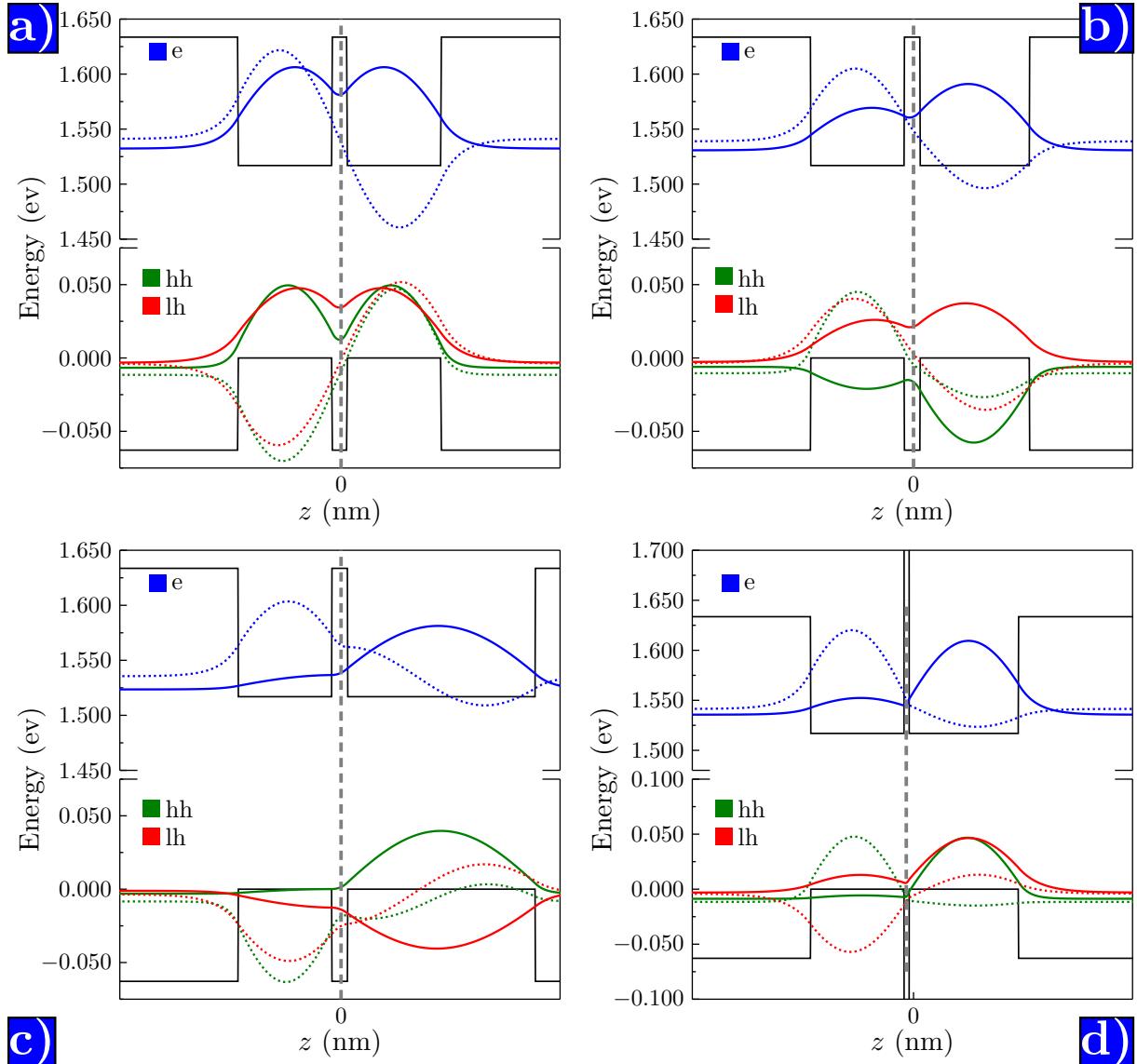


Figure 2.10: From (a) to (d) shcemes the numerical results obtained to solved 1D-Schrödinger equation in both SQWs and ACQWs.

The barrier width as can see plays an important role, because the width define tunneling of electrons and holes, for example Figure 2.10(b) and Figure 2.10(d) structures has the same wells widths with exception to barrier type, the first one is Al_{0.15}Ga_{0.9}As and the width is $b_w = 1.98\text{nm}$ while the second one is AlAs type with a width of $b_w = 0.565\text{nm}$. It's known that the difference between these is the bandgap energy then, as is expected, the tunneling is less than the Al_{0.15}Ga_{0.9}As barrier. Concerning tunneling, if compares the Figure 2.10(a) and Figure 2.10(c) structures, which basically are the same structures except for the width of the second well, the wave function in one of these, are symmetrically localized as a doublets states, while the other the wave function practically is well localized in one of the

wells as single state energy of a single QW [26]. While if compares with Figure 2.10(b) where one of the coupled wells is slightly width than the other, the wave function distribution seems an SQCQWs under applied external electric field [88], later in Section 3.2.2 this take a sense, in PR experiments appear transitions which we associated to three-body particles known as trions (\mathbf{X}^- or \mathbf{X}^+). Trions are particular particles that consist of the bound of two electrons and one hole (\mathbf{X}^-) or vice versa, two holes and one electron (\mathbf{X}^+). The fact of one of the wells is slightly width, entails quantum interactions measured experimentally. But, this doesn't only the unique, interesting phenomena observed in these structures. As before mentioned this work, focuses on the optical properties consequent of the structural asymmetry, this means, the relative width in double wells.

2.5

Anisotropy model in CQWs

Now, we focus on the central part either, the core of this work. The asymmetry in structure entails a very interesting quantum process in these semiconductor structures. The symmetry reduction is the basis of the model in this work, as explained before, symmetry is the cause of many quantum phenomena its exhibits in solids, and the fact, that symmetry breaking in CQWs increases the optical anisotropy, which among many properties this opens the field of spin properties [88, 120–123].

Generalizing the before discussed in the symmetry section exists several mechanisms to reduce symmetry in QWs structures, principally in the single QWs with external perturbations such as electric or magnetic field, mechanics perturbations as applied strain among others. But, the objective is the same, reduce the symmetry in these structures from T_d which is the symmetry group to cubic crystals to C_{2v} which in turn a subgroup of this. As the result of the symmetry breaking, it originates an OA which was measured through RAS experiments results in a peculiar way, this refers to the spectral results. In our case, it compares the experimental results of RAS experiments (later discussed) about three CQWs structures, in which the difference between these is only the width of one of the wells (as many times mentioned), the RAS spectra increase as the relative width also, this, come back to these structures very interesting. Therefore, it will discuss the Anisotropy Model which includes all of the previous contained and focused on the CQWs. The IOA was borns as of the Quantum-mechanically effect of mixing between holes states, the theoretical formalism frequently is developed around effective mass but it's important to remember the difficulty of solid physics, now if it adds the physics of the interface this is so crazy. The role of interfaces is crucially in heterostructures in point of fact is the principal cause, then, having a standard model which explains it, is practically impossible. Therefore the *simplicity method is ever the good choice*.

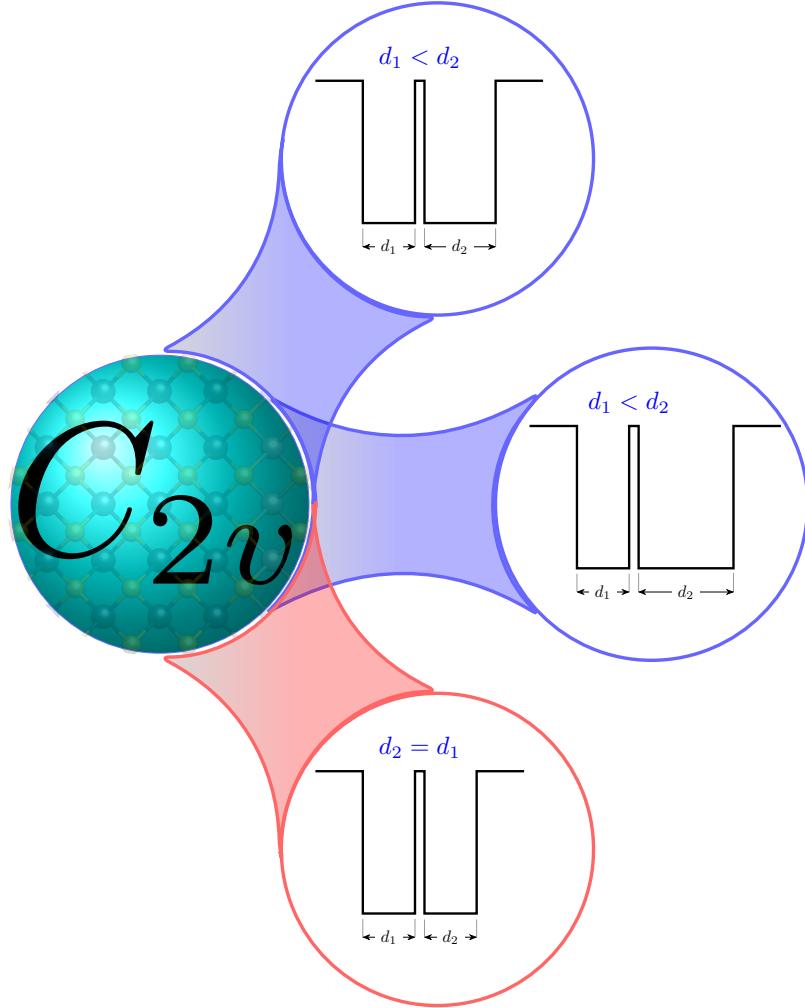


Figure 2.11: Diagram of three principal structures studied in this work, started with CQWs with the same width (bottom, $d_1 = d_2$), this sample structure is called as SCQWs, the second one the first asymmetric structure (top, $d_1 < d_2$), this means that one of the wells is slightly width than the other. Finally the third sample (right) it's more asymmetric, which means that one of the wells is double wider than the other.

The EFA usually employs the analytical method to explain the heavy- and light- holes states lack of interface contribution [124]. In the fact, always it desires high-quality interfaces but even though the growth technologies are very precise and powerful this remains a natural atom process. It's important to say that the quality of structures studied it's amazing, even in the SCQWs were expected a non-observable anisotropy due to the conserve of D_{2d} symmetry, experimentally observes a remanent of anisotropy. As it's exposed in the Figure 2.11 the three CQWs structures studied, including the symmetric double wells sample which remarks in red because this is the basis to demonstrate the evolution of anisotropy as increases as the asymmetry in wells also to. If we inside into discussion we increase the uncertainty of who is the best method to explain the IOA increases, on the one hand, the perturbative methods like the $\mathbf{k} \cdot \mathbf{p}$ have a powerful several Hamiltonians while

the “atomistic” as TB which advantage is the consideration symmetry intrinsically. Then as mentioned before we employs one method based on EFA, which consider each interfaz a scalar potential [71, 75, 124, 125]. Then, to deal measure the hole-mixing caused by interface anisotropy it’s important that this be sensitive to changes in polarization, by this reason, the models by a QW’s consider a hole-mixing Hamiltonian H' as [75, 124, 126]:

$$H' = \{E_p + [P_0\delta(z - w/2) - P_0\delta(z + w/2)]\} \{\hat{J}_x \hat{J}_y\} \quad (2.10)$$

with

$$\{\hat{J}_x \hat{J}_y\} = \begin{pmatrix} 0 & i & 0 & 0 \\ -i & 0 & 0 & 0 \\ 0 & 0 & 0 & i \\ 0 & 0 & -i & 0 \end{pmatrix}. \quad (2.11)$$

The Equation (2.10) is employs in a SQW [75], here, we denoted E_p as the external perturbations parameter, this parameter usually involves the multiplication of electric field along z direction, D as deformation potential and other as the piezo-electric constant d_{14} . The δ is the “abrupt” interface as a function of position z and well width w and P_0 is the potential parameter, finally considers the angular momentum operators with basis [75]:

$$\left| \frac{3}{2}, \frac{3}{2} \right\rangle, \left| \frac{3}{2}, -\frac{1}{2} \right\rangle, \left| \frac{3}{2}, \frac{1}{2} \right\rangle, \text{ and } \left| \frac{3}{2}, -\frac{3}{2} \right\rangle \quad (2.12)$$

This Hamiltonian is usually implemented to explain the interface anisotropy, of course, doesn’t only limit to SQW, also is implemented to superlattice structures [126], where is considered a summation of interfaces contribution. In our case, this Hamiltonian H' it’s not enough to explain the hole mixing which origins the OA, the principal reason is that this H' considers the interface sign \pm refers to BA or AB interfaces, then the total mixing would be canceled by that reason. Also, this last is not necessarily true, remember that this would occur in an ideal interface where is expected a structure with D_{2d} symmetry then the IOA should be zero. Then, the non-abrupt interfaces should be generated a residual IOA [19, 75, 125]. RAS spectra of single QWs have been reported extensively in the literature [12, 16, 18, 19, 75].

The thickness of the QW plays a fundamental role in the amplitude of the RAS signal. For the anisotropies originating at the interfaces and the ones induced by the segregation of Ga or In (in the case of the inclusion of a thin InAs layer), the RAS amplitude decreases when the thickness of the QW increases [75]. Contrary to this behavior, for the IOA induced by strain, the RAS signal increases with the thickness of the QW [18]. In any case, the RAS

signal is associated with the mixing between the heavy- and light-hole valence subbands of the QW [71]. In a perturbative approach, the IOA strength is proportional to

$$\frac{\langle \psi_{\text{en}} | \psi_{\text{hhn}} \rangle \langle \psi_{\text{hhn}} | \mathcal{H} | \psi_{\text{lhn}} \rangle \langle \psi_{\text{lhn}} | \psi_{\text{en}} \rangle}{\Delta E_n}, \quad (2.13)$$

where $\langle \psi_{\text{en}} | \psi_{\text{hhn}} \rangle$ and $\langle \psi_{\text{lhn}} | \psi_{\text{en}} \rangle$ are the overlap integrals between the $|\psi_{\text{en}}\rangle$ electron state in the conduction band and the $|\psi_{\text{hhn}}\rangle$ and $|\psi_{\text{lhn}}\rangle$ hole states in the valence band respectively. The mixing between heavy- and light-hole subbands is $\langle \psi_{\text{hhn}} | \mathcal{H} | \psi_{\text{lhn}} \rangle$, \mathcal{H} being the perturbative Hamiltonian. The difference in energy between the hole states before their mixing is $\Delta E_n = E_{\text{hhn}} - E_{\text{lhn}}$.

As previously mentioned, \mathcal{H} could account for a perturbation by applied uniaxial stresses, electric fields (built-in or external) and abrupt or smooth interfaces. In the case of CQWs system, \mathcal{H} is a measure of the asymmetry of the two QWs. In a CQWs, if both QWs have the same thickness, the electron and hole energy levels are brought into resonance and their probability density is distributed symmetrically at both sides of the barrier that connects both QW, as can be seen in Figure 2.10. If the AlGaAs/GaAs interfaces of the DQW are equivalent the symmetry of the DQW structure belongs to the D_{2d} crystallographic point group. However, as we mentioned, if the GaAs/AlGaAs interfaces (sources of anisotropies) are non-equivalent, and a residual IOA is induced by the symmetry reduction from D_{2d} to C_{2v} of the whole CQWs structure. In this case the number of symmetry operations is reduced to four: the operations that inverts the z -axis, are excluded, thus leading to a mixing of heavy- and light- holes in the valence band, and hence an IOA. When the thickness of one QW is larger than the other QW (ACQWs), the energy of the electron and hole states of each QW become different and are no longer in resonance. In this case, the probability density is distributed asymmetrically as can be seen in Figure 2.12(a). Besides of the anisotropic source induced by the GaAs/AlGaAs interfaces as in the symmetric case, for the asymmetric CQWs system another mechanism of IOA should be considered. Suppose that the AlGaAs/GaAs interfaces are equivalent. As we mentioned, in this case a symmetric CQWs system belongs to the D_{2d} symmetry. If the thickness of one QW is modified, the system changes symmetry from D_{2d} to C_{2v} . As can be seen in Figure 2.12(b), the symmetry operations that change the sign of the z -axis (i.e., a reflection on a plane perpendicular to z), are no longer possible because the AlGaAs/GaAs interfaces are located at different distances from the barrier. The number of symmetry operations of the ACQWs structure is reduced to the four operations of the symmetry group C_{2v} [73, 76]. In this case the IOA strength can be modulated by changing the relative thicknesses of the QWs in the CQWs system. This anisotropy is added to the residual IOA produced by the non-equivalence of the interfaces.

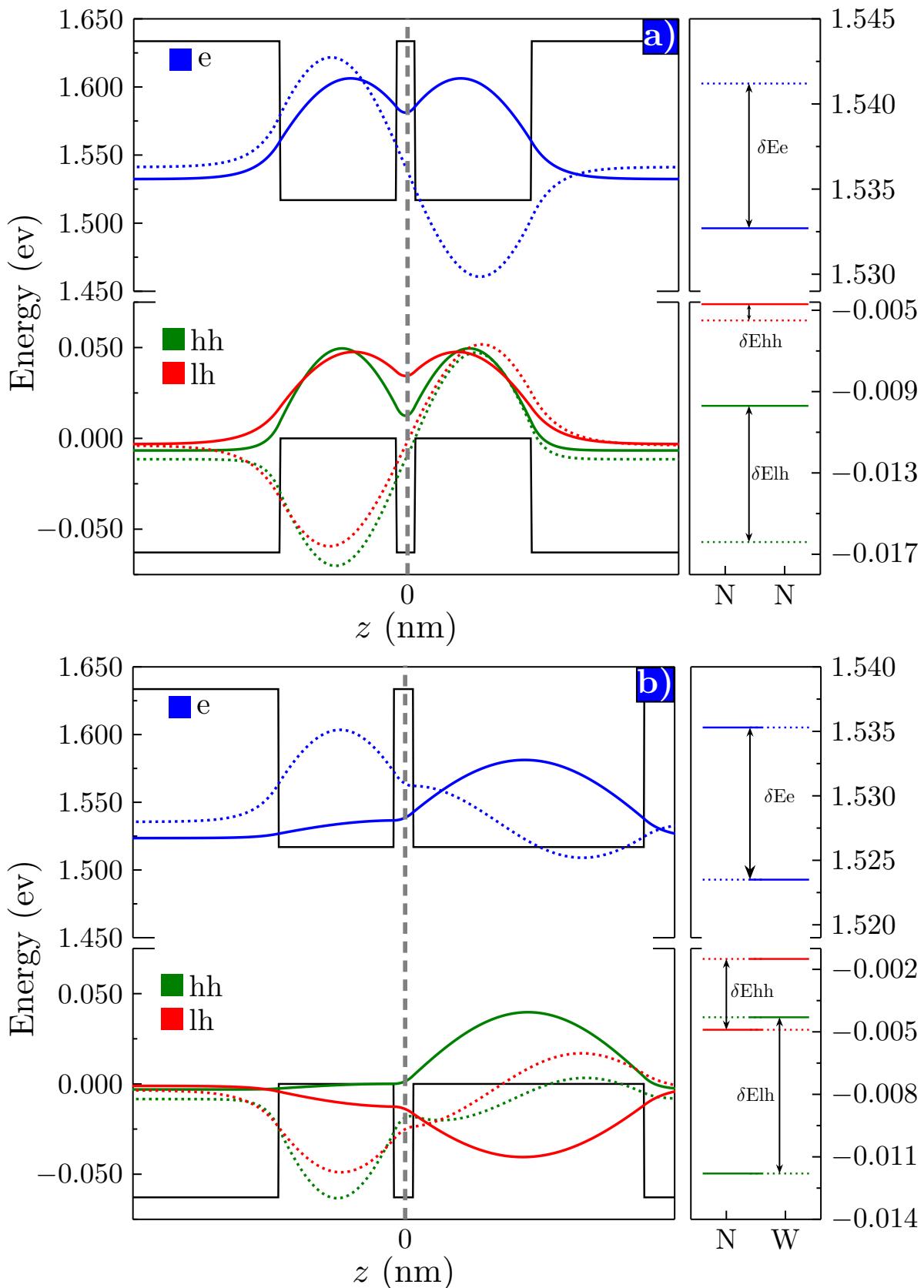


Figure 2.12: Wave functions and confined energies calculated for (a) sample S1 (SCQWs sample) and (b) sample A2 (ACQWs sample 2)

2.6 Summary

In this chapter explains and discusses the physics which involves this work, started from relation between symmetry and the bandstructure in semiconductors, the importance which understand the symmetry context and the importance of the “breaking” in it (symmetry reduction). We focus on enhance the particular symmetry “breaking” in CQWs which occurs practically natural, this means, in this work we don’t appeal to apply and external perturbation to success.

3

EXPERIMENTAL DETAILS AND RESULTS

This chapter shows the experimental results in the spectroscopy detailed in it which are compared and correlated among them, to success a physical explanation of the obtained results.

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3.1 Samples Description

QUANTUM STRUCTURES were created thanks to the technic Molecular Beam Epitaxy (MBE) growth which in general terms consist in great precision of the growing thin films of semiconductor materials, through high precision deposition on a suitable crystalline substrate. As has been pointed out on many occasions, MBE is nothing more than a sophisticated form of vacuum evaporation* [127], this does not sound bad but the MBE technique is not only this, epitaxial growth needed the precision in experimental growth parameters how temperature, deposition rate calculations, REED analysis, etc. So that in a bit of words the purpose is to supply appropriate atoms or molecules to the substrate surface and leave surface diffusion, surface reactions and inevitable desorption from the surface to play their various roles in generating an epitaxial film [127, 128].

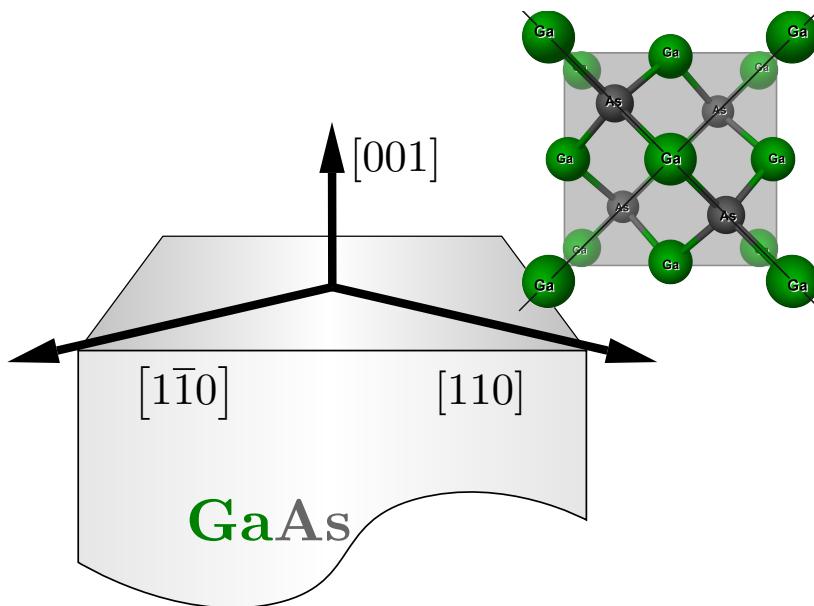


Figure 3.1: GaAs substrate

The samples studied in this work, are made up of a GaAs (001) substrate, the GaAs substrate has a zincblende-type crystal structure, in which the atoms are tetrahedrally coordinated. Figure 3.1 the choice of crystal is very important and preparation of these can modify the growth result, for this reason, the high quality of substrate is the basis of growth of heterostructures. The heterostructures are a combination of more than one semiconductor, this leads to the superlattice definition where more of two semiconductors are

*In owner experience, MBE growth is a very exhausting task, so great respect to all people that works on this

atomically deposited over the substrate in the growth direction (001). The semiconductors used in the samples are GaAs and AlAs then these can have different thicknesses, growing periodicashowgrid][lly and alternately way over the substrate with thickness $d_{\text{GaAs}} + d_{\text{AlAs}}$. The result consists in an arrangement of $(\text{GaAs})_2(\text{AlAs})_2$ superlattice, like a shown in Figure 3.2 or AB arrangement like a said commonly. In QWs structures, the width is the principal characteristic because quantum confinement is dependent on this, then MBE is the perfect choice to realize this.

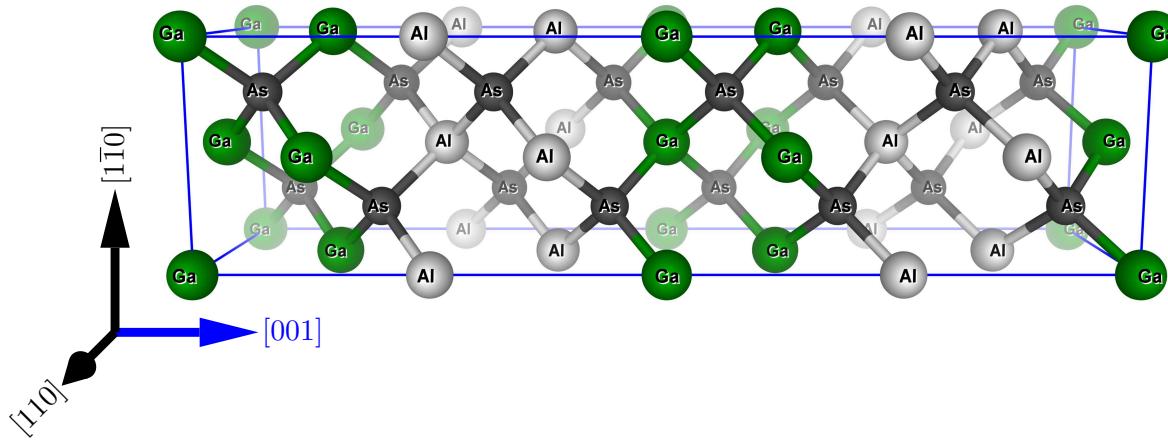


Figure 3.2: Atomic order in AlGaAs crystal structure grown along (001) direction.

The realistic structures present some details that can modify the experimental results, some of these are inevitability due to the complexity of growth. One of these is the interfaces between two different materials, i.e., the typical structure of GaAs/AlGaAs has an interface between these, then the problem is due to an imperfect mismatch of materials, therefore, can cause anisotropy effects. Other details which usually are considered are surface, point defects along of structure, or strained caused by overlayer in the epitaxial process. Later, the interfacial detail gives the physical sense to RAS experiments in the symmetric CQWs structures.

Table 3.1 shows the sample names, each sample has two QWs coupled by a thin barrier preferentially of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ semiconductor where Al percent is important because of the barriers potentials depends on this, in the case of AlAs barriers the potential barrier is bigger than $\text{Al}_x\text{Ga}_{1-x}\text{As}$. As a before-mentioned, this was taken into account in numerical calculations to generate the potential profile. Commonly for the case, $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers both in adjacent like the coupling barrier the x values are in the interval $0.1 < x < 0.4$.

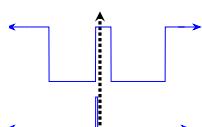
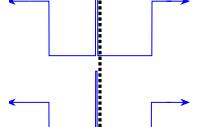
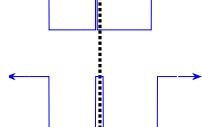
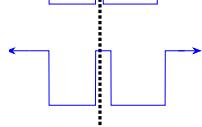
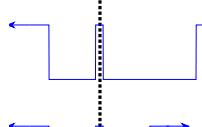
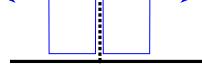
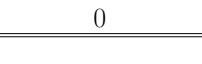
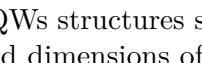
Sample	NW width (nm)	$V(z)$	WW width (nm)	Barrier width (nm)	Barrier	Adjacent barriers	Doped type	$\left[\frac{1}{cm^3} \right]$
M4_3171	11.87		13.85	3.960	AlGaAs	Al _{0.15} Ga _{0.85} As	n[Si]-i-n[Si]	6×10^{18}
M4_3172	11.87		13.85	0.565	AlAs	Al _{0.30} Ga _{0.70} As	n[Si]-i-n[Si]	6×10^{18}
M4_3226	11.87		13.85	0.424	AlAs	Al _{0.30} Ga _{0.70} As	n[Si]-i-n[Si]	6×10^{18}
M4_3140 (ACQWs-1)	11.87		13.85	1.980	AlGaAs	Al _{0.15} Ga _{0.85} As	i-n[Si]	6×10^{18}
M4_3141	11.87		13.85	3.960	AlGaAs	Al _{0.15} Ga _{0.85} As	i-n[Si]	6×10^{18}
M4_3521 (ACQWs-2)	11.87		23.74	1.980	AlGaAs	Al _{0.15} Ga _{0.85} As	i-n[Si]	6×10^{18}
M4_3522 (ACQWs-3)	11.87		23.74	1.980	AlGaAs	Al _{0.15} Ga _{0.85} As	i-p[Be]	5×10^{16}
M4_3523 (SCQWs)	11.87		11.87	1.980	AlGaAs	Al _{0.15} Ga _{0.85} As	i-n[Si]	6×10^{18}

Table 3.1: This table shows the CQWs structures studied in this work. CQWs potential profiles $V(z)$ are shown to observe the different shapes, composition parameters, and dimensions of structures studied. The dashed line determines the symmetric reference in the last samples in which we focused (M4_3141 , M4_3521 , M4_3522 , M4_3523), due to their characteristic results.

Worth mentioning that the purpose of the first four samples (up to down in table) they were grown with the objective of measure indirect excitons (IX), so that, the structures are more complex, and we omitted the value of n-doped since these samples are doping in bottom and top to minimize the intrinsic electric field (screening field effect) so that external perturbation is applied through a voltage over the sample [129] and the n-doped enhance the external perturbation *.

After realized experiments over the four first samples and observe possible trions (X^+ or X^-) through PR spectroscopy and an apparent increase of the RAS signal, it was decided to focus the experiments on these structures, which have a less complex composition as in the case of the first three samples in the table (see Table 3.1). Therefore, it took the sample M4_3141 as a basis, i.e., same barriers widths both coupling barrier and adjacent barriers, obtained the samples M4_3521 , M4_3522 and M4_3523 where sample M4_3522 is the same to sample M4_3521 in structure but with a different type of doping (p-type in M4_3522 and n-type in M4_3521), samples M4_3521 and M4_3522 have the same doping type (n-type in both) but one of the QW is more width to another, this means that the sample M4_3523 have the same thickness in both QWs (SCQWs) and sample M4_3521 have one QW thick more than the another (ACQWs).

The Section 3.2 shows each experimental setup implemented in this investigation with their respective results. Starting with the Section 3.2.1 shows PL spectroscopy, their experimental setup implemented to obtain transitions energies values and compare with numerical results, after, in Section 3.2.2 shown experimental setup and results of the PR spectroscopy, to get information about the intrinsic electric field and the effects caused by this. Finally, Section 3.2.3 shows the RAS experimental setup and exposes the experiments that were realized to study the anisotropy caused by the asymmetry due to the relative thickness between the CQWs.

3.2

Spectroscopy: Experimental setups and results

OPTICAL SPECTROSCOPY frequently is defined as a branch of physics that studies the *light-matter interaction*, this is important because the simple definition of interaction covers a vast realm of physical phenomena from classical to quantum electrodynamics [130]. Therefore, optical spectroscopy is an essential tool in experimental solid state physics, gives the guide to study optical and electronic properties of semiconductors.

The optical process in semiconductors consists of the study of response due to *light-matter*

*Non-published, the specific information about the doped in these growths, if you have a question about this, you can send email to Dr. Klaus Biermann biermann@pdi-berlin.de

interaction, this response corresponds basically to the processes that can occur in the solids when light* falls on (photons) in it. These processes are absorption, reflectance, emission, and scatter where all of this depends on electromagnetic spectra range, in our case this range includes from near-infrared to mid-infrared (700nm to 900nm). Although those processes are of the utmost importance, optical spectroscopies that uses in this work involve more interest in absorption and emission, being the latter sensed in our experimental setups.

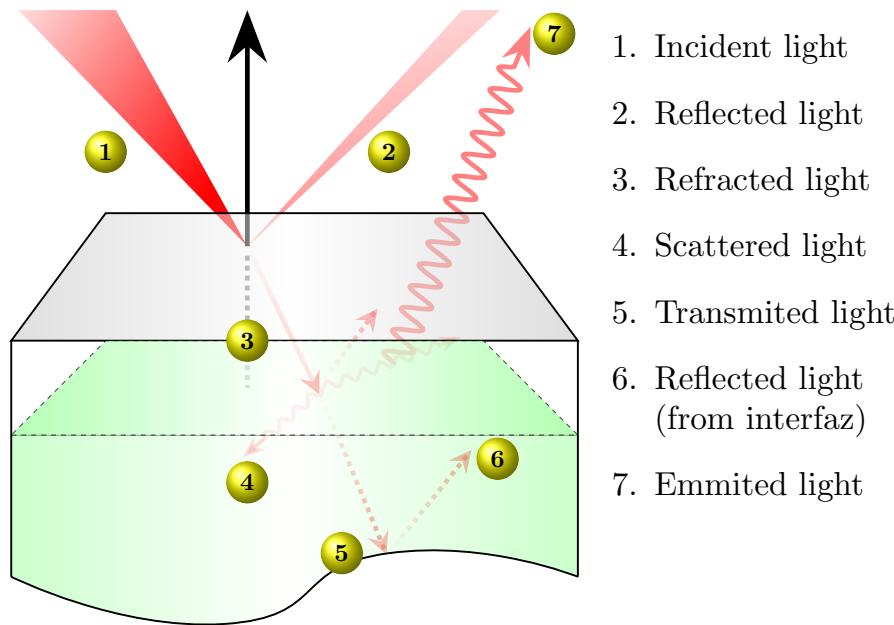


Figure 3.3: Processes that occur inside a solid in the light-matter interaction phenomena.

Before continuing with the manuscript of this chapter, I would like to mention that the experiments could not be carried out without the development of detectors, specifically optical detectors. To speak about optical detectors, it is impossible not to mention the photoelectric effect that in general terms is the transfer of energy from photons to electrons when the light spot on a surface [131]. So, this fundamental quantum phenomenon is the basis of detectors in spectroscopy, these detectors are called photodetectors, where these convert the light power(photon incidence over their sense area) into an electric signal, voltage, or current, then this signal we can measure and amplify it. The next sections will mention and discuss the PD implemented in the experimental setup and especially which are the pros and cons. It is important that the photo-cathode of the photo-detector corresponds to the spectral range of experiments. i.e, in our experiments the spectral range of interest is from 700nm to 900nm the PD should have characteristics to adequate of this range. The Table 3.2 shows some PD and their characteristics [29].

As a comment, it is important to denote that the CCD devices are been span in many

*Refers to light due to radiation spectral range

Cathode	Range (nm)	$\phi(\%)$	$\lambda(\text{nm})$	$i_d \text{ nA}$
bialkali (S-22)	300-630	26	400	0.1
multialkali (S-20)	180-800	20	480	0.2
extended red multialkali (S-25)	300-900	7	600	1
GaAs	300-920	15	700	2
Cs-Te	160-320	14	200	0.01

Table 3.2: Photo-cathodes, usually implemented in PD to the spectroscopy of semiconductors [29].

setups in recent years and this is because these devices enlarged the range of experiments that can be realized with these, above all in other areas of physics as in the experimental astrophysics in which has obtained awfully important results. Nowadays, CCD devices in experimental solid-state physics have contributed to getting great experiments that previously were limited due to spatial resolution and time response. In the section 3.2.1, it discusses the advantages and cons of the detectors (PD and CCD) in PL spectroscopy context.

3.2.1 Photoluminescence Spectroscopy (PL)

Photoluminescence spectroscopy is characterized by be a fast spectroscopy to get optical properties (i.e band-gap) and transitions in semiconductor materials, for this reason the work began with PL spectroscopy with aim of searching optical transitions in each CQWs samples and compare with numerical solution of one-dimensional Schrödinger equation (see Table 2.1).

Although PL signal is characterized by be greater than other spectroscopies implemented in this work, the need to use a chopper is only to filter the signal to the external noise, this is achieved used a lock-in amplifier where the reference signal is the chopper and signal input is first measured by a multimeter and then input in the lock-in amplifier. In many other experimental setups the experimental measure are take fast, this is because implement a CCD device as a detector of experimental signals, where these devices are distinguished by fast time of acquisition (apart from other reasons). In our case the time of measure it does not comparable with those, due the time in our experiments is about 2 hours (explained latter) and those are about several minutes. But why use lock-in amplifier if CCD devices shorter measured time?

The answer is not be for impatient*, the reason is that, the lock-in and detector implementation allow control over spectral resolution through two parameters, the first one is in the choice of monochromator's slits apertures and the second one has to do with step

*The author self-considered impatient

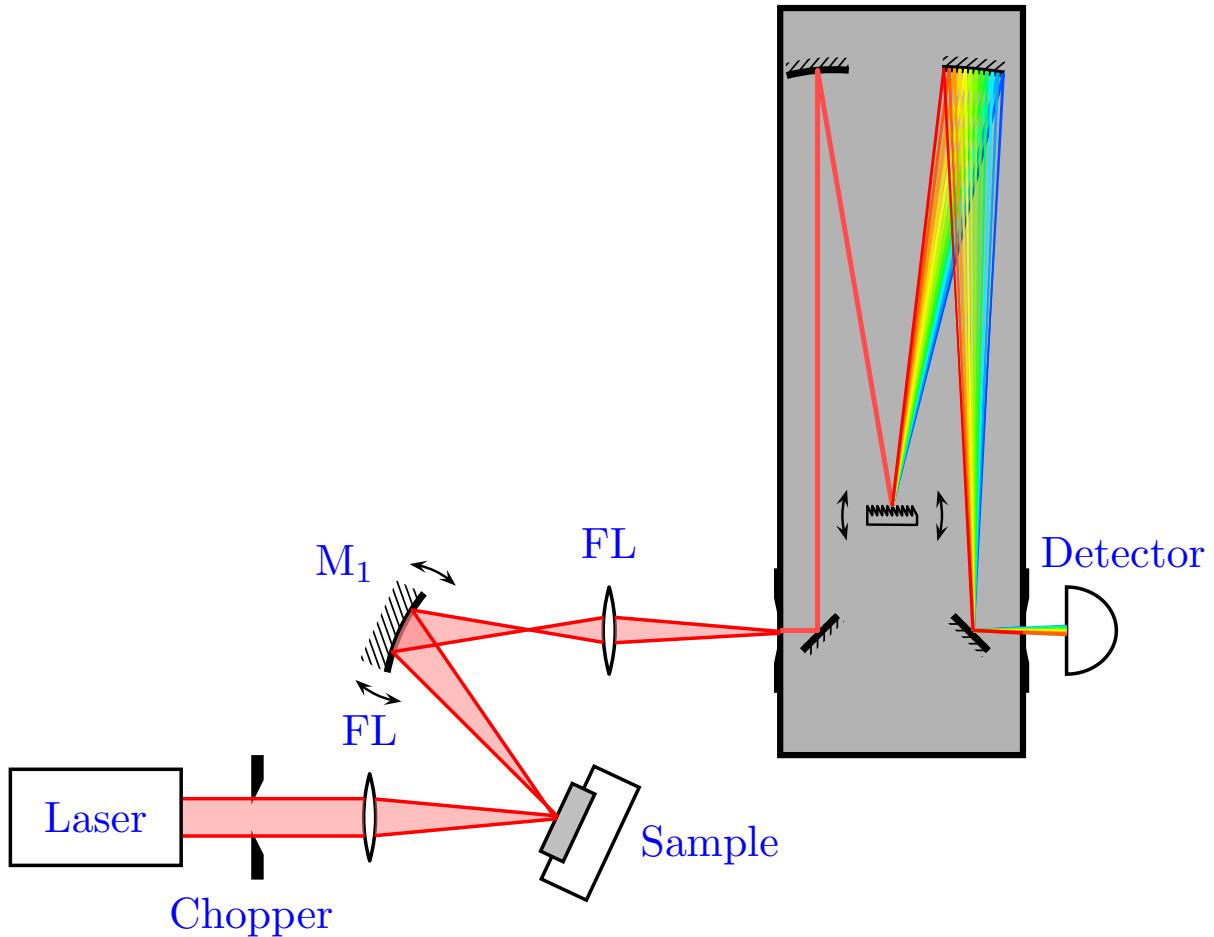


Figure 3.4: Scheme of photoluminiscense setup, the temperature of experiments at 14 K, the wavelength of laser 685nm and was used a Si detector.

wavelength this is the progress step by step over the spectral range taking into account in the experimental measure. Also, the step by step experiments provide the choice of measures number in each step where finally only consider the average of these points, resulting more clean spectra in compare with CCD setups*. Although the time is important worth it inverts to get quality experiments.

Some experiments shown below correspond to Carlos's bachelor thesis [132], who implemented a simple and reliable PL system also he makes a computational code to fit PL spectra using numerical and experimental results (you can check Carlos's codes in our laboratory repository on GitHub[†]). These results were correlated with previously realized experiments taking into account the same parameters.

The experiments are organized by labels, how shows in Table 3.1, started with the samples

*Even if modify time and average of measures, the spectra measured with lock-in amplifier are more quality.

[†]<https://github.com/Spectroscopies-Lab-IICO>

M4_3171 , M4_3172 and M4_3226 where these samples were grown with objective the measure trions. The experimental parameters are shown in Table 3.3, in each experiment the optical setup was optimized to enhance the signal measured, the optimization process consisted in measure laser peak at a definite monochromator slit's aperture then finely move the mirror until achieving a high response in multimeter, repeat this closing the slit's and measuring the FWHM of the laser peak in each step, finally we obtained an optimal resolution about 1 nm in our PL experiments.

Sample	Laser	Range(nm)	λ step (nm)	No. of singnal acquisition	Slits aperature (μm)
M4_3171	680	800-820	0.1	20	75
M4_3172	680	780-840	0.1	15	75
M4_3226	680	800-820	0.1	20	100
M4_3140	680	800-820	0.1	20	100
M4_3141	680	800-820	0.1	20	100
M4_3521	680	800-820	0.1	20	100
M4_3522	680	800-820	0.1	20	100
M4_3523	680	800-820	0.1	20	100

Table 3.3: PL experimental parameters implemented in each sample, all experiments were carried about 14K and was used the same red (680 nm) laser diode. The measured parameters as a Wavelength step or number of acquisitions per step Wavelength were optimized as explains in the text.

An important detail about module laser ThorLabs and very remarkable in the Section 3.2.2 is related to power, the power of this was not stable in some time periods causing a variation in the results, this is the reason to use different experimental parameters and after some test were optimized the choice of the Wavelength step, number of acquisitions per λ step and slits aperture, this depending on each sample.

The PL spectra, corresponding to the samples showing in Figures 3.5(a) and 3.5(b), were more complicated to interpret and in experimental conditions as well. For this reason, we decided to calculate the absorption along of the structure that composes each sample because due to they have more layers. We speculated that this is the reason why it was complex to realize the experiments and their analysis. As previously spoken, the study of light-matter interaction in solids can be a headache this is because in real experiments more than one interaction mechanism can be observed, especially in PL spectroscopy and, the objective is to measure only one of them, therefore, the experimental results can be affected and complicate their interpretation. The reason for starting with the samples of Figure 3.5 is that they have effects that modify the PL spectra and, although these effects are very interested in our case, they reduce the PL signal.

If started with the scoop, that the PL spectrum in semiconductors is given by an interband emission generated by recombination carriers, and this, in turn, is due to the absorption

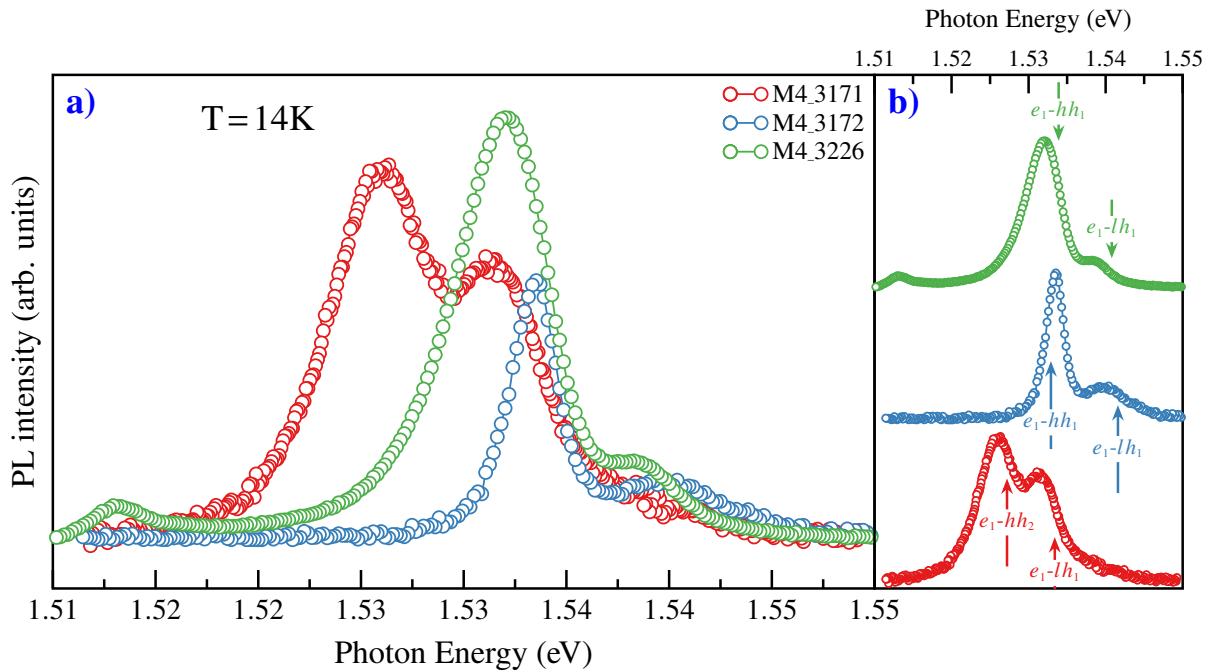


Figure 3.5: Subfigure (a) shows PL experiments of the samples M4_3171 , M4_3172 and M4_3226 , where show spectra and results measured at 14K. Plots in how the comparison between these samples, where clearly can see the relative intensity among in each experiment. In (b) it can be seen each PL spectra with the respective direct transitions numerically calculated, hh1 and lh1 indicates the first energy level that corresponds to heavy- and light-holes respectively.

of photons provided by an excitation laser source. Therefore, the PL signal is due to absorption, it does not matter that they are opposite processes, the absorption, as well as many other mechanisms of light through the solid, can modify and generate the spectrum of PL. The absorption analysis was carried on in the macroscopic approach to light-matter interaction (in Chapter 4, talks about quantum processes and aspects of light-matter interaction in a microscopic environment), therefore, can use classical electrodynamics to study optical properties of semiconductors. One crucial property of semiconductors is the dielectric function, this is proportional to the complex refractive index (later it is named only as refractive index) and with this, can be described the optical properties of semiconductors. It is important to mention that the refractive index describes how light propagates in a medium, and it expressed as [41, 133]:

$$\tilde{n} = n + i\kappa = \sqrt{\varepsilon(\omega)} \quad (3.1)$$

where the real part (n) represents the refractive index and the imaginary part (κ) is the extinction coefficient. Even if the objective of this work does not rewrite and reinterpret the physics of these phenomena, we will try to focus on specific equations to reach the

goal, which is the absorption model in semiconductors. Then the electric field inside a solid can be written as [133–135]:

$$\mathbf{E} = \mathbf{E}_0 \exp\left(\frac{-\kappa\omega z}{c}\right) \exp\left(-i\omega\left(\frac{nz}{c} - t\right)\right). \quad (3.2)$$

In the Equation (3.2), can see the solution of Maxwell equation in a macroscopic picture of the photon-material interactions. This represents a wave propagating with dispersion and staying in terms of refractive index, but the principal idea is to establish a relationship that can help us to describe the absorption in terms of these principal parameters. The absorption is a process that occurs in any spectroscopy and is the absorption coefficient that defines such a feature in each medium. The extinction coefficient κ is the cause of the wave damping when the electromagnetic wave crosses in media, therefore it's related by absorption coefficient. Now in Figure 3.3 can see that there are three principal parts, that they are intensities that are reflected, transmitted, and absorbed. These three parts compose the original incident light through:

$$I_0 = I_R + I_T + I_A \quad (3.3)$$

Finally, we call upon one of the most notable laws in spectroscopy, this basic expression relates the intensity of light absorbed with the absorption coefficient and is known as Beer-Lambert law [136, 137]:

$$I(z) = I_0 \cdot e^{-\alpha z}, \quad (3.4)$$

then this tell us how intensity decrease as a function of absorption coefficient. As a result of these basic principles in a real structure, we can define de absorption in each layer. After these general and quick basics of the macroscopic basis of light-matter interaction, let's move to a more realistic environment where the calculations are more complex and where it is decided that parameters have major physical priority. The numerical solutions of absorption along structure were carried out using an exceptional Open-Source code called **SOLCORE** [6]* and our codes. As previously mentioned in a real calculation is frequently taken into account only parameters with major physical sense depending on the situation, arduous computational solutions, or for simplicity. From **SOLCORE**, we have the simplest model to calculate the absorption, despising all reflections at the interfaces having only the absorption as a function of wavelength and depth z expressed as:

$$A_n(\lambda, z) = \alpha_n(\lambda) \exp\left(-\sum_{i=1}^{n-1} \alpha_i(\lambda) d_i - \alpha_n(\lambda)(z - z_n)\right) \quad (3.5)$$

*You can test and contribute this code, visiting their GitHub repository at <https://github.com/qpv-research-group/solcore5.git>.

where α_n is the absorption of layer n , d_i is the thickness and z_n the position of beginning of the layer*.

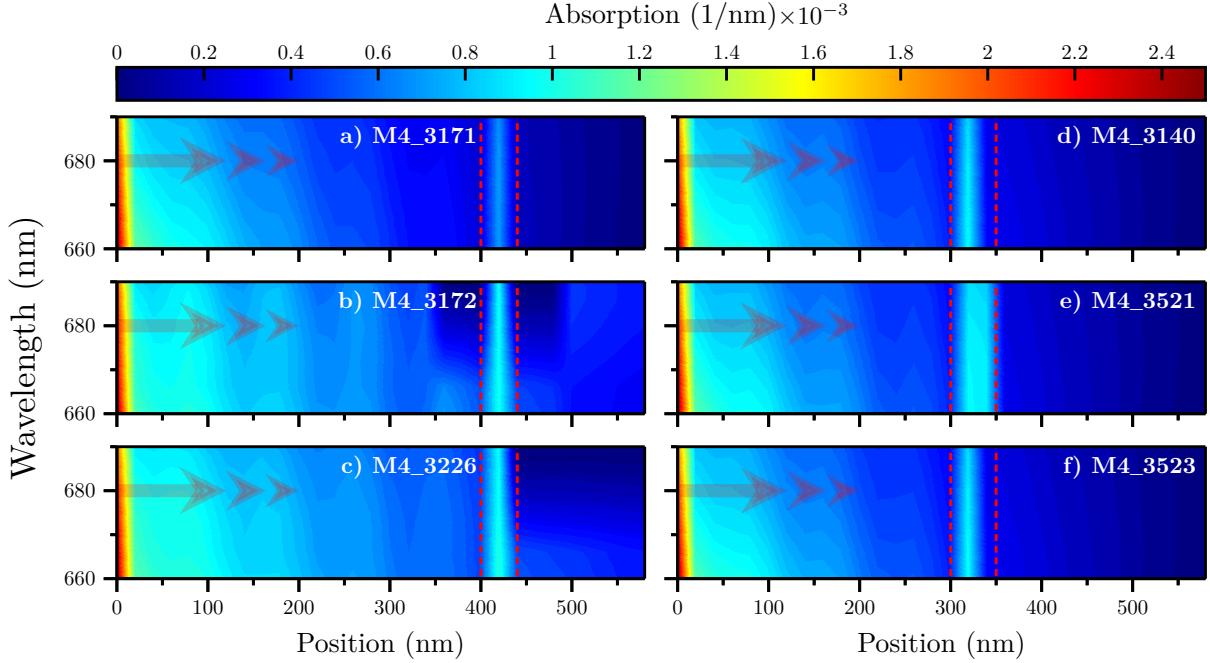


Figure 3.6: Absorption calculated as a function of sample depth, dashed lines closed the CQWs region, left: Figures 3.6(a) to 3.6(c), here the samples have $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers with different compositions $x = 0.15, x = 0.2$ and $x = 0.3$ this results in a change of refractive index then also expected in absorption. Right: Figures 3.6(d) to 3.6(f) these samples are equal in structure but change the width of one oM4a)f the QWs, therefore the absorption manifests more homogenous than the first three samples.

The samples with extra layers have characteristics that don't present in samples with fewer layers, one of this is which the fundamental transition around the GaAs gap doesn't possible to observe in these samples, the amplitude in these samples in comparison with sample M4_3141 for example, is around of twenty times smaller than this, opposite case in samples M4_3140, M4_3421, M4_3523. We take these samples because these are the basis of this work, the complete set of PL experiments corresponds to the work previously mentioned. So, why is the important to get the PL results and calculate absorption? The answers are simple, the importance to get PL spectra to help us to get experimentally energy transitions, which have a very important role in the next experiments and in the basis of our model to explain the increase in anisotropy in the ACQWs. Also, the experimental PL results are the basis to compare that our numerical calculations are consistent with the experiments. Numerical calculations get more complicated in large structures, this means, while the structure is conformed with more layers and these layers

*In eq. (3.5) originally the thickness is represented how w_i , by confusion issues it was decided to change the notation.

are thicker, the probability to get a divergence in calculations is great, this is the reason which not all models work. In fact, in our numerical calculations the energy transitions in samples with more layers (M4_3171, M4_3172, M4_3226) we had to be careful at the moment to choice energy binding, the reasons are many and in the PL is complicated to get unique information and even less in structures with high doped, the charge recombination they play us dirty to understand the results.

The case of absorption calculations is a guide to explain the difficulty to carried out PL experiments and put to discussion if doping is a reason to generate or not intrinsic electric field, after all the absorption is part of field solutions.

Figures 3.6(a) to 3.6(c) are the calculations of absorptions as a function of depth, where previously explained, it was only taken into account classical regime therefore the absorption remains in terms of the optical parameters of each layer which conforms all structure. In these figures can shows that in a range of 650 nm to 700 nm the wavelengths are absorbed with major proportion in the first layer, this is due to the structures starts with a GaAs doped(n- or p-type) or undoped layer. After along in structures, also can observe the decrease absorption to short wavelengths, and the notably increased around of the coupled quantum wells.

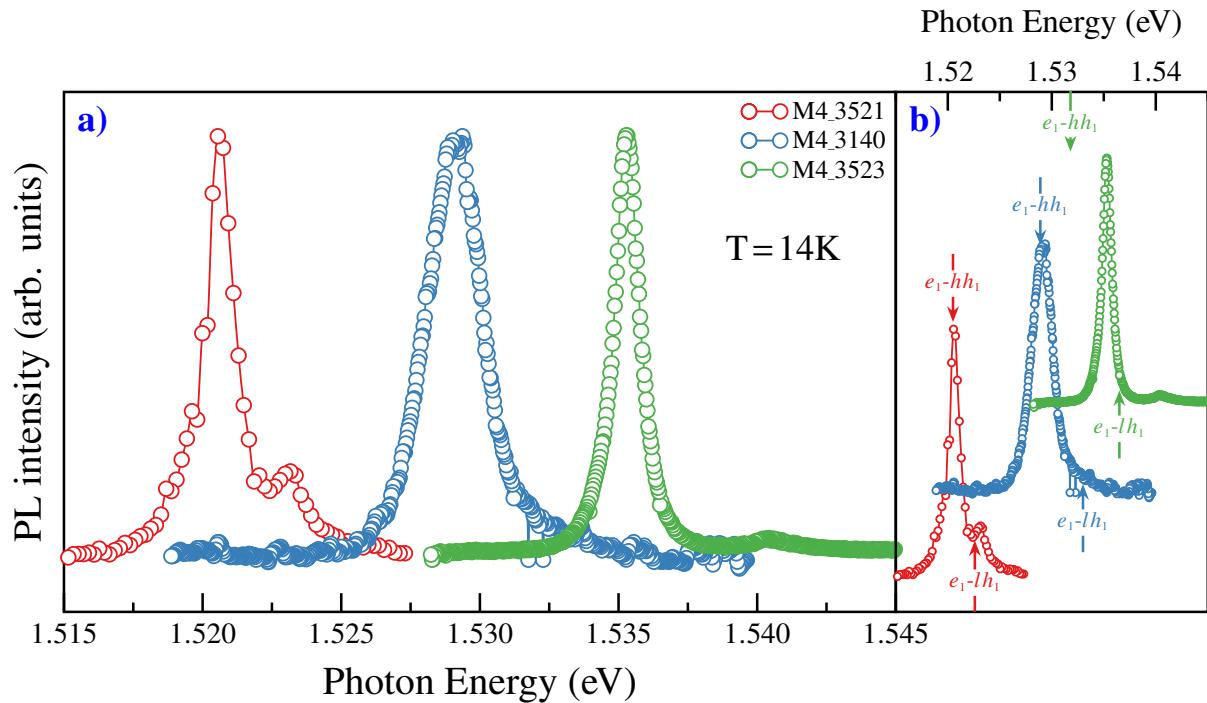


Figure 3.7: (a) Shows the PL spectra to samples M4_3140, M4_3521 and M4_3523, in this comparison is clear the shift between these in respect to first transitions. The relative change in width of one of the QW modify the energy transitions being the sample 3521 the lowest energy. (b) It is plotted each PL spectra result with the correspondent e_1-hh_1 and e_1-lh_1 transitions energies.

These calculations make sense in the next section where PR spectroscopy is a powerful tool that can be used to measure the optical properties due to the modulation without external perturbation, this means that the modulation depends on the intrinsic properties of the structure. The generalizability of the results is limited by the classical regime, as previously mentioned, it has taken into account the refractive index where the absorption can obtain from the complex part of this, that is the extinction coefficient. The common use of the PL is to determine the band gap and optical transitions in semiconductors, especially in QS as quantum wells, but what happens if consider PL regardless of the penetration depth? In our case it is important doesn't, due to the structures studied have wide layers before of coupled quantum wells region, this can observe in figure 4 where shows the results of samples: M4_3171, M4_3172, M4_3226, M4_3140, M4_3521, and M4_3523. In the PL experiments carried out here, it was used as a show in Figure 3.4 a red laser diode of $\lambda = 685$ nm, although the laser energy is chosen in respect to energy gap therefore short wavelengths are preferable but how we can see these wavelengths are absorbed in the first layer due to the samples ends in a GaAs layer, even the laser used is absorbed in large proportion.

Contrary to the samples measured first, the samples M4_3140, M4_3521, and M4_3526 exhibits the most spectral resolution this means that exhibit a more homogenous behavior due to the QWs have quantized energy, therefore, the electron-hole recombination is more probably to measure than the other samples which present width spectrums so that these results may be due to the several internal mechanisms of which can be: impurities, large carrier density due doped, defects, among others [138, 139]. In each of these PL spectra, Figures 3.5 and 3.7 present the most intense peaks associates with heavy- and light-holes exciton transitions denoted by e1-hh1 and e1-lh1 respectively.

Transition	M4_3171	M4_3172	M4_3226	M4_3140	M4_3521	M4_3523
e1-hh1	(E)	1.5270	1.5341	1.5329	1.5170	1.5207
	(N)	1.5313	1.5342	1.5339	1.5292	1.5196
e1-lh1	(E)	1.5318	1.5402	1.5286	1.5234	1.5406
	(N)	1.5370	1.5414	1.5408	1.5335	1.5220

Table 3.4: Comparison table between experimental transitions obtained through PL measures and numerical transitions calculated as explained in Section 2.3

In discussion with the before mentioned mentioned, several mechanisms can contribute to getting an inhomogeneous spectrum, even it can say that the two peaks which correspond to exciton transitions are thick and merge this can be related with the high doped level this is because very high dopant concentration causes an overlap of the impurity band with the free-carrier continuum [139]. Table 5 shows the comparison between experimental transitions energies get with PL and the numerical results. It is important to mentioned

that the approximation of numerical calculations are closed to experimental, the difference is about of 5 meV for the PL case. It is well-known that the PL signal is increase as a function of decrease well widths, as shows

Table 3.4 shows the comparison between experimental transitions energies get with PL and the numerical results. It is important to mention that the approximation of numerical calculations is closed to experimental, the difference is about 5 meV for the PL case. It is well-known that the PL signal is increased as a function of decrease well widths, this due to the energy of the confined particle state depends on strongly in it and this is demonstrated in Figure 3.7 this due to structures doesn't have top n-type epitaxial layer these structures are i-n type, staying only barriers structures and PL line shape presents strong confinement [140–142].

3.2.2 Photoreflectance spectroscopy (PR)

POTOREFLECTANCE belongs to the group of modulation spectroscopy, being one of the most important to determines field effects without external perturbations, this means that there is no need for an external source that generates the electric field on the sample. Exists another's kinds of modulation spectroscopy, where the type of modulation depends on interest effects, these can be phenomena linked with temperature (thermoreflectance), strain (piezoreflectance), electric (electroreflectance), etc. There is a great information amount of based on this technique, so it decides as previously mentioned and not to repeat in future chapters or sections, the principal idea is to focus on more representative expressions and phenomenological interpretation which be the best following our models and results. Unlike the PL the PR is the reflectance measure as a function of modulation or the changes in it, this once more needs to involve optical properties of the sample studied, in the eq. (3.1) is expressed the refractive index with their real and imaginary part respectively and these are proportional to dielectric function. If the PR is the change in R which is due to modulation of an intrinsic electric field generated by doped layer o layers in the sample (later discuss this mechanism) this mean that:

$$\frac{\Delta R}{R} = \frac{R_{\text{off}} - R_{\text{on}}}{R_{\text{off}}} \quad (3.6)$$

where R_{off} and R_{on} are the reflectivity when the perturbation (laser) are activate or not are the reflectivity when the perturbation (laser) are activated or not, this mean that the results falls on perturbation that is the laser.

The PR as modulation spectroscopy is a powerful tool to perform the study of semiconductors, their modulation mechanism occurs when the built-in field is screening by photoexcited carriers created through incident photons, which involves contactless and

non-destructive. In many cases, this spectroscopy technique is preferred due to it can measure transitions in heterostructures at room temperature in comparison with the PL or PLE [143] that are measured at low temperature. Therefore, the highlighter characterize of the PR is the modulation of the built-in electric field, in part, this is due to the structure characteristics but in fact, the PR spectra is the change that generated electric field in the dielectric function, this is expected because is the result of measured the changes in reflectivity generated by the laser, in other words, the laser induces an excess of carriers which neutralize intrinsic field. This is well-known to study in the bulk materials, the models show as the reflectivity change is very well approximated by a first-derivative [144–147] but in QWs structures that are dominates by excitonic transitions then the PR line shapes can be understood in terms of modulation of dielectric function appropriate for excitons [143, 148–150].

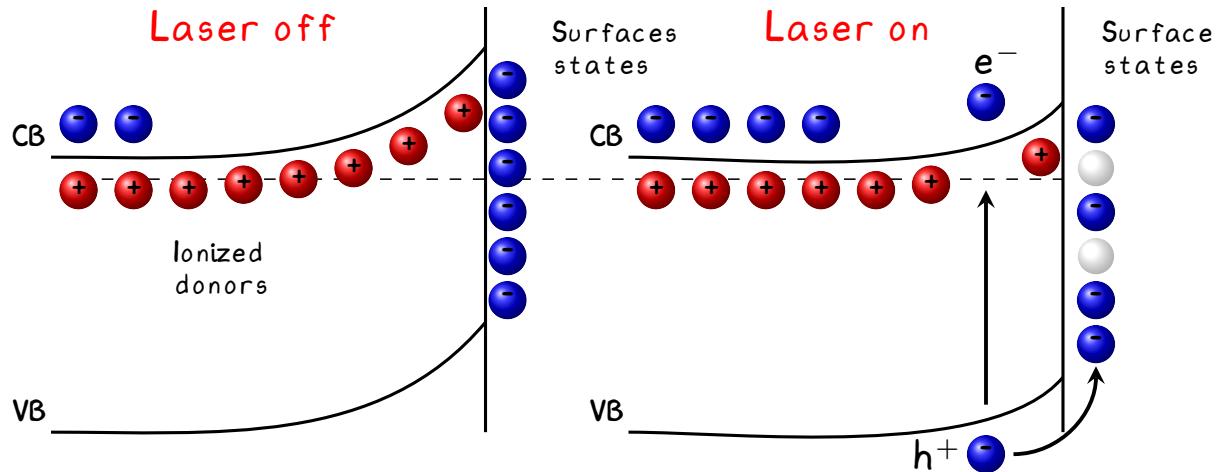


Figure 3.8: Scheme of the PR effect where it shows the carrier dynamics, in left can see the photoinduced changes by the laser is applied.

Under the objective of this thesis, we have not focused on doing a traditional study of the line shape with their respective model, the study over PR lineshape is complex, the interpretation starting from changes in dielectric function like a derivative shape and depending on magnitude perturbation, i.e, doesn't is same the PR analysis on structures with free carriers in solid that the confine particles in QWs structures, the bounded particles are not accelerated by built-in field modulation, so the energy spectrum is discrete and not continuous like free particles. The capability to implement a lineshape fit, do the PR be a great tool. It has been mentioned which the photoreflectance process is due to the built-in electric field modulation, therefore we are considering that the sample was grown considering desired characteristics to generate an intrinsic field, this means that sample contains n-type doped layers, impurities, unintentional strain mechanisms which generate a space charge region. In the case of an n-type doped layer in a structure, create a space charge region, this region creates the field therefore the conduction and valence bands are bending as a show in the Figure 3.8 and the Fermi energy are pinning at the surface.

Photoexcited electron-hole pairs are separated by the built-in field, with the minority carrier (holes in this case) being swept towards the surface.

At the surface, the holes neutralize the trapped charge, reducing the built-in field [146]. That is the general explanation to the PR modulation, before we mentioned that a characteristic of the PR, is to fit as a derivative-like and the order depends on structure, also, we mentioned which in quantum wells the confinement and bound states modify the line shape and the respective fit. So, in this situation, the modulation of field causes the binding energy change of excitons, in other words, this is a Stark effect but in an inverse case because of the field it already exists. The electron-hole pairs depend on binding energy, if that is modified, then the intensity of the transition varies.

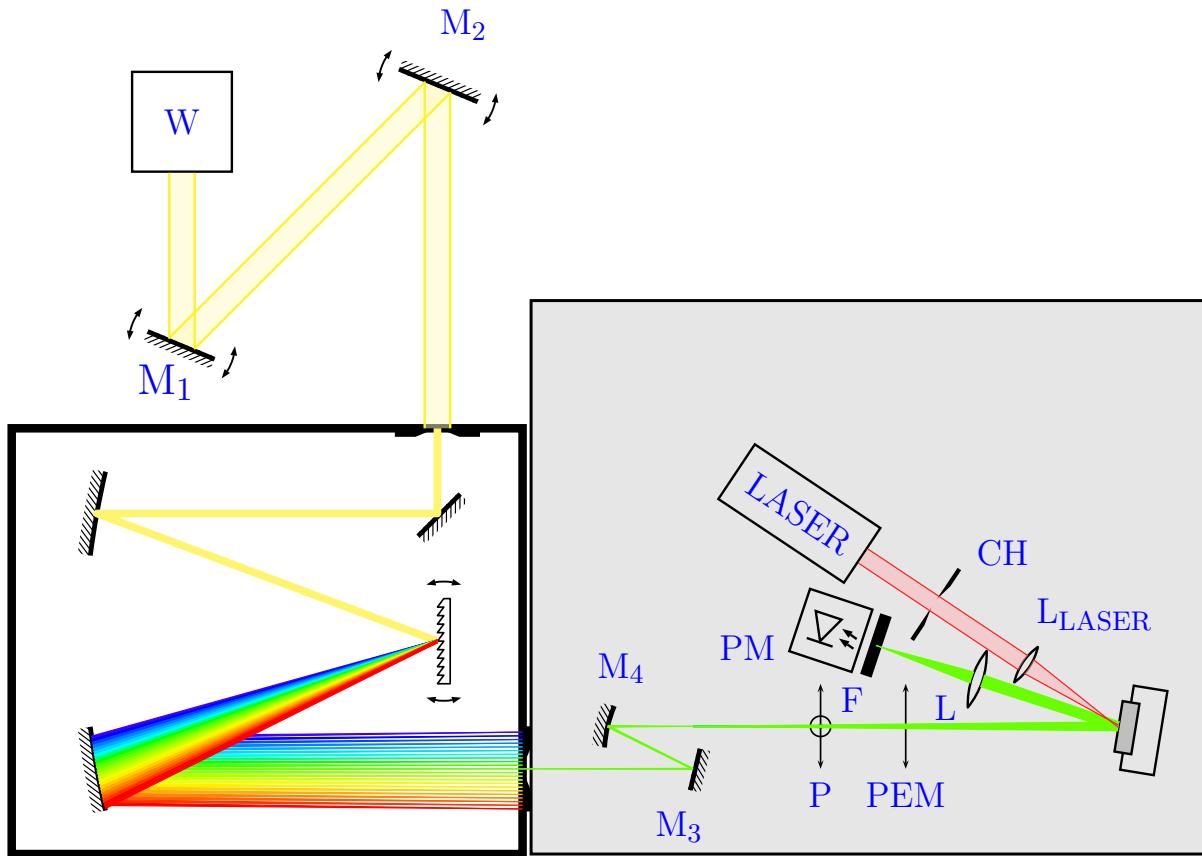


Figure 3.9: Photoreflectance setup used in these experiments, the setup implemented is commonly called dark configuration this due that photo-detector are exposed, then keeping closed to ambient light. W: tungsten lamp, M₁ to M₄ variable mirrors, P: polarizer, PEM: photoelastic modulator, L: focus lens, F: filter, PM: photomultiplier, L_{LASER}: focus lens for laser, CH: mechanical chopper.

The experimental setup to PR experiments implemented in this work, shown in Figure 3.9. The setup start with the probe light from a tungsten lamp, the beam is led by two silver mirrors to monochromator entrance slit, then the monochromatic beam passes through a polarizer and a photoelastic modulator finally affects on sample. The reflected light is focus to the PM with a focus lens. Modulation of the electric field in the sample is caused by photo-excited electron-hole pairs created by the pump source in our case is a red laser

that illuminates the same spot of the monochromatic beam and is chopped to a certain frequency, in this setup we use a mechanical chopper at 1KHz. It is important to mention that the reflected light in addition to being focused by a lens, is filtered before incide at PEM, this is important because the reflected laser light by the sample can modify the modulated R signal, do not forget that the PL signal is involved too.

Although the R signal is modulated by the chopper at 1KHz, after is measure by a lock-in amplifier due to the change in the R is very small about of 1×10^{-4} , in comparison with the PL signal, R change is less than PL as one million times. This is the reason by which any modulated spectroscopy commonly uses a lock-in amplifier. In our case, the setup is called Dark [146] setup because the PM is exposed to room light, therefore the system is keep closed. The Dark configuration has some advantages, one of these is, that the R changes are subtracted intrinsically therefore the use of the filter is enough to the dispersion of laser isn't a problem. We refer intrinsically to the R signal modulation. If the PL signal achieves to be detected, the system will perform subtraction as shown in Equation (3.6), if the PL signal mixes with the R in both cases this will cancel because it is constant, staying only the change in R.

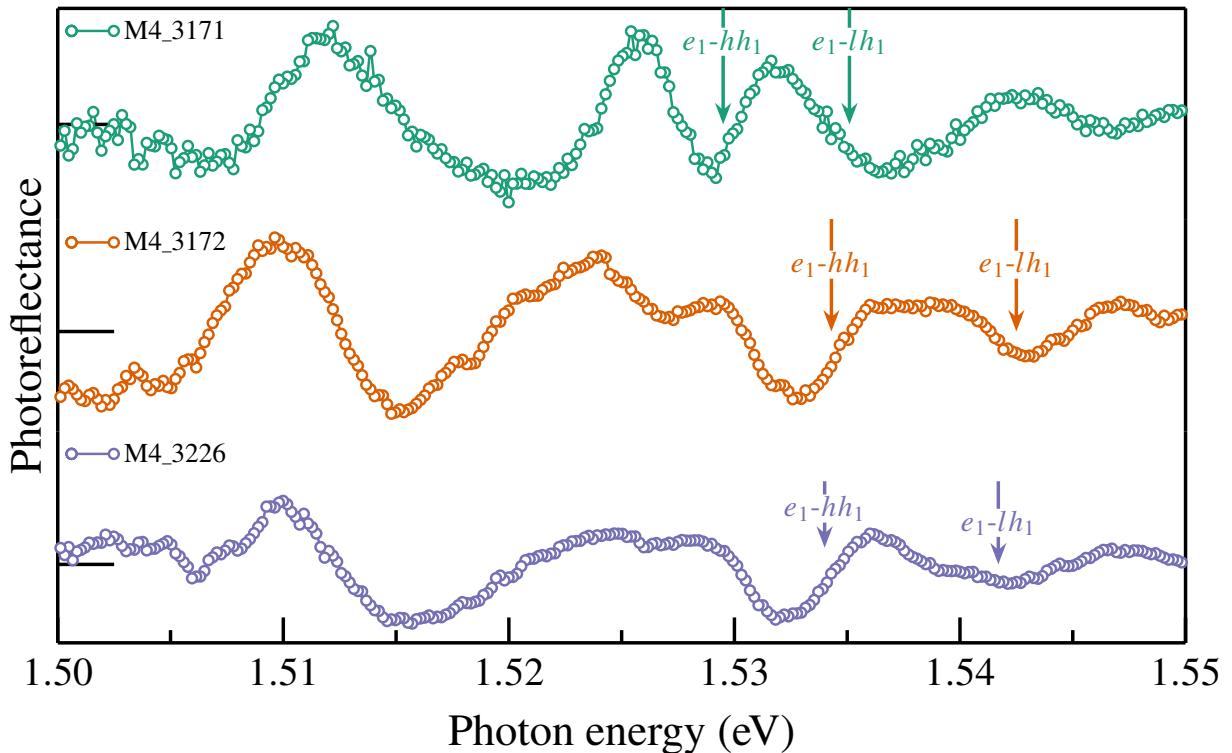


Figure 3.10: PR experiments to samples: M4_3171, M4_3172 and M4_3226 at 30K. Arrows point to calculated transitions for each sample, the used laser wavelength was the same, which in PL experiments and the power used in each of these was 5mW. Dashed line point the GaAs substrate.

The result of PR signals associated with band to band and quantum level transitions,

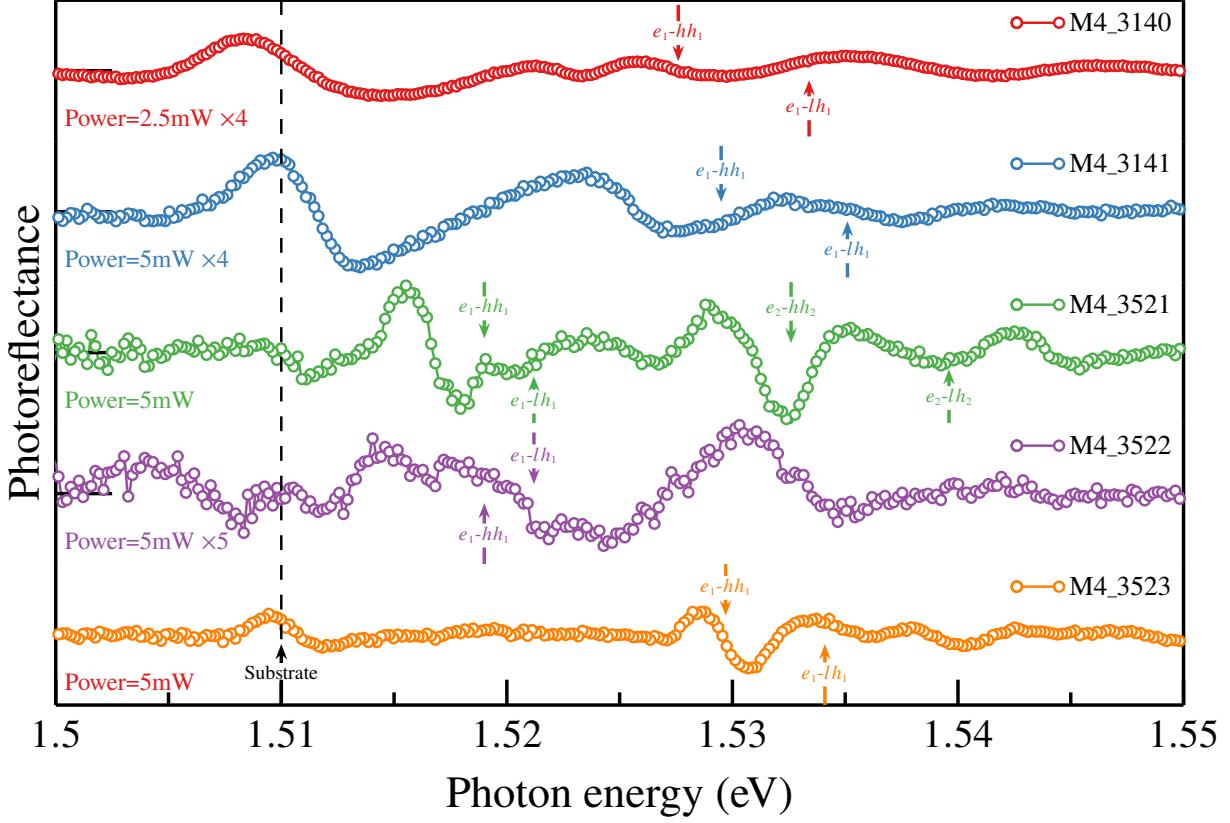


Figure 3.11: PR experiments to samples: M4_3140, M4_3141, M4_3521, M4_3522 and M4_3523 at 30K. Arrows point to calculated transitions for each sample, the used laser wavelength was the same, which in PL experiments and the previous PR experiments. Dashed line indicate the GaAs substrate, in these samples this transition is well located. The PR spectra of sample M4_3522 p-type doped (orange) is five times smaller than their correspond n-type, the sample M4_3521. The experiments in sample M4_3140 was performed at 2.5mW, then was multiply it by four in according to samples M4_3521 and M4_3523, in case of the sample M4_3141 even if, was performed at a power = 5mW the result was 5 times smaller than sample M4_3521. The discussion about of this is explained in the text.

in case of samples M4_43171, M4_43172 and M4_43226 the built-in electric field is low due to the structure n-i-n, this in principle is canceled or screen, this mean that the field create by carriers is opposite. The Figure 3.10 shows the result of PR experiments on samples mentioned above and are a bit informative. In fact, GaAs gap isn't visible in these samples, many mechanisms can affect the PR results in these. The first one and more representative undoubtedly is the low built-in field, if the structures are n-i-n the field expected is so low although the field can affect the interfaces and contribute but in general this is cancel by opposite photo-carrier directions. Frequently the PR is used tool to calculate or estimate intrinsic fields, this is possible in intermediate-field regimen, this known as Franz-Keldysh oscillations (FKOs) by electric field along z -direction [151]. So, the in PR spectra is observed oscillations and the period is determined by the field in the structure, where typically only about 3-4 FKO can be detected in the space charge region

of a doped sample. In this case for these samples and as will be explained later, the PR experiments are complex in context to determine all transitions that occur in them and the field is smaller, therefore does not have FKO.

The samples n-i-n type as shown in the Figure 3.10 the FKO does not exist and this is clearly observable because these experiments does not have any oscillations, as before mentioned this behavior may be occurred because the directions of photocarriers generated are opposite, then the intrinsic field is canceled. Thus, the PR measured spectra are the result of modulation of intrinsic residual electric field or nonuniform fields effects [152]. In contrast, with the n-i-n type samples, the i-n type samples exposes clearly the direct transitions even associated to transitions with more energy (next levels energies), although the intrinsic field is not enough to generate FKO.

In the Figure 3.11 can see the direct transitions numerically calculated for samples M4_3140, M4_3141, M4_3521, M4_3522 and M4_3523. The sample M4_3521 was taken as reference in terms of amplitude, in the sample M4_3140 was performed experiments as a function of power laser, being the power 2.5 mW the closest at 5mW, this is one of the reasons for that the result spectra was approx four times smaller than the sample M4_3521. The sample M4_3522 p-type, maybe can be five times smaller than their n-type analogous sample (M4_3521), but the line shape resultant does not have any response, i.e., in terms of amplitude is smaller, but the transitions aren't clarified or not resolved. This is one of our keys to final results because it has to do with carrier distribution and the nonexistence of the built-in electric field. For the sample M4_3141 the PR spectra is similar to the sample M4_3140, the difference between these structures is the barrier width, but in spectra the transitions are more resolved in the sample M4_3141.

From now on, we focus on the samples : M4_3140, M4_3521, M4_3522 and M4_3523, these samples are similar in structure, if shows experiments of the n-i-n type samples and the rest of i-n type samples is only to remark the importance of our results.

Even though in many works about the PR is normal to submit a line shape fit model to clarify the effects of modulated intrinsic fields around of critical points or transitions and as before mentioned this doesn't the interest of this work. Although, the study and models of line shape for modulated spectroscopy as the PR are essential in the experimental study of semiconductor physics [153, 154]. Nevertheless, in our experiments although the PR mechanism is observable in general around of the Gap (E_0) and direct transitions, the mechanism of modulation over the low intrinsic field, exhibit effects which is not frequently in the PR experiments, as it's shown in the following section.

3.2.2.1 Excitonic effects

THE CQWs structures are useful to study excitonic effects under external perturbations of an applied electric field. These structures are coupled by the thin barrier, then the electrons overlap over both wells, this behavior is very studied by the confinement effects and possibility to create devices based on electron properties. In this work, we focus on the samples which exhibit exciton effects that commonly are observed under external field apply in our case, without external fields or external perturbations and nor any structure modifies*. Starting with a comparison between ACQWs n- and p-type these are the M4_3521 and M4_3522 samples respectively, Figure 3.12(a) shows the PR of both samples(left), where the p-type sample is approx five times smaller than the n-type sample, therefore we can conclude that the mechanism of photo-carriers generated in these are different even if it has the same CQWs structure.

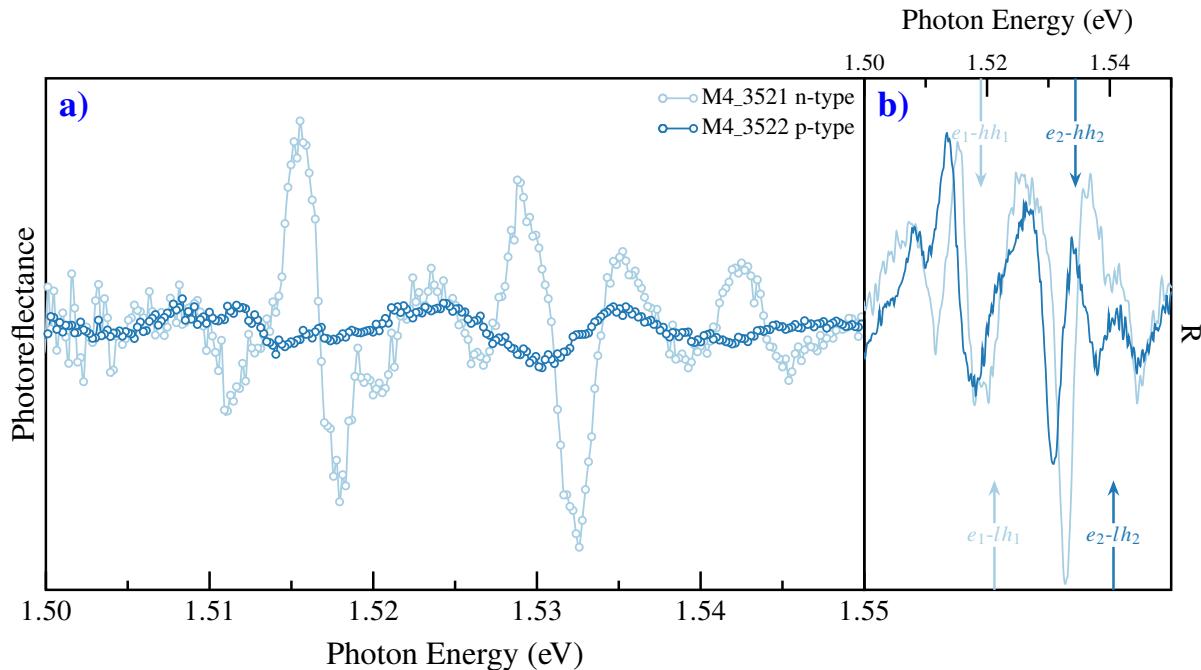


Figure 3.12: Comparison of : a) PR spectra of the 3521 (n-type) and 3522 (p-type) samples, the p-type sample is 5 times smaller than n-type sample. b) R spectra obtained at same time in each experiment, the arrows point to each direct transitions for two first confined energies. The line shape is practically the same in both spectra.

Also, the R spectra as shows the Figure 3.12(b) obtained synchronously at each experimental measured, this is the DC signal detected by photomultiplier and reading by the multimeter. These signals are practically the same, according to equation eq. (3.6) the change originated

*This refers to create strain by polish over the samples or mechanisms which generates strain, external perturbations also refers to temperature changes, applied currents, and others.

by the laser source doesn't enough, i.e., $\Delta R = R_{\text{off}} - R_{\text{on}}$ is smaller. In another way, the comparison of samples M4_3521 (ACQWs-2) and M4_3523 (SCQWs), shown in the figure Figure 3.13 exhibits the difference in amplitude at same power laser but around de E_0 is well resolved in both. In case of the SCQWs sample the direct transitions are well resolved, but in ACQWs-2 sample, could be present forbidden transitions, pointed at Figure 3.13 with same color of its correspond PR spectra. This behavior it has been observed previously [155] in MQWs structures even at 300K [156]. The nomenclature to allowed transitions (direct transitions) is: en-hhm for electron-heavy hole and en-lhm for electron-light hole, n index represent the n-th conduction subband and m-th valence subband. So, when n=m its refer to direct transitions or allowed transitions, in ACQWs appear peaks related to transitions between first electron energy where electron wave function is predominantly in the wide well, but this wave function is overlapping to narrow well even if in minor percent, this mean that n \neq m then the heavy- and light-holes confined at narrow well can create e1-hh2, e1-lh2 transitions, or the electrons in second confined energy that predominantly are at narrow well but, they can penetrate (tunneling) to the another well (wide well) as seen in Figure 3.13 can generate another forbidden transitions. It's important to mentioned that this behavior is presented at low-field regime, therefore can't associate this to modulation of the built-in electric field, this being discussed since some years ago [156, 157], even though can this associated, with the behavior which have the electron, heavy- and light-holes in CQWs structures, with a specific barrier width and the height of the potential barriers (or depth of wells) [26, 158]. The electron and holes tunneling depends on those parameters and, in our case, the barriers potential depends on Al percent in the alloy $\text{Al}_x\text{Ga}_{1-x}\text{As}$, therefore $x = 0.15$ then the barriers they are not so tall, and the coupling barrier width is very thin (< 2 nm).

These structures, become an interesting platform to study the phenomenology and behavior of confined electrons, heavy- and light-holes, and their respective interactions. Another interesting phenomenon, which we could observe, was the triions (X^+ or X^-) formation through the PR experiments. When were carried out the set of PR experiments over the samples i-n type, occurred a peculiar event while we performed and established the correct measure parameters, to be specific while we determined the laser power. This doesn't mean that our experiments are wrong, as a matter of fact, this peculiar event made us test the experimental setup several times and carried out experiments as a function of laser power. This event started with the M4_3140 (ACQWs-1) sample, this sample has a well with a slightly wide width and because of that the wave function overlapping in major percent than the others samples, by this reason is reasonable or expected, that the triions formation it's more likely as explain later. While they were being carried out, the PR experiments in ACQWs-1 sample, at higher power laser allowed by our device, in fact, this was trouble, because the laser power doesn't stable, so it was decided to turn on the laser previously before performing each experiment, this was around of eight hours before to start experiments. After detecting the problem with the laser power, it was started

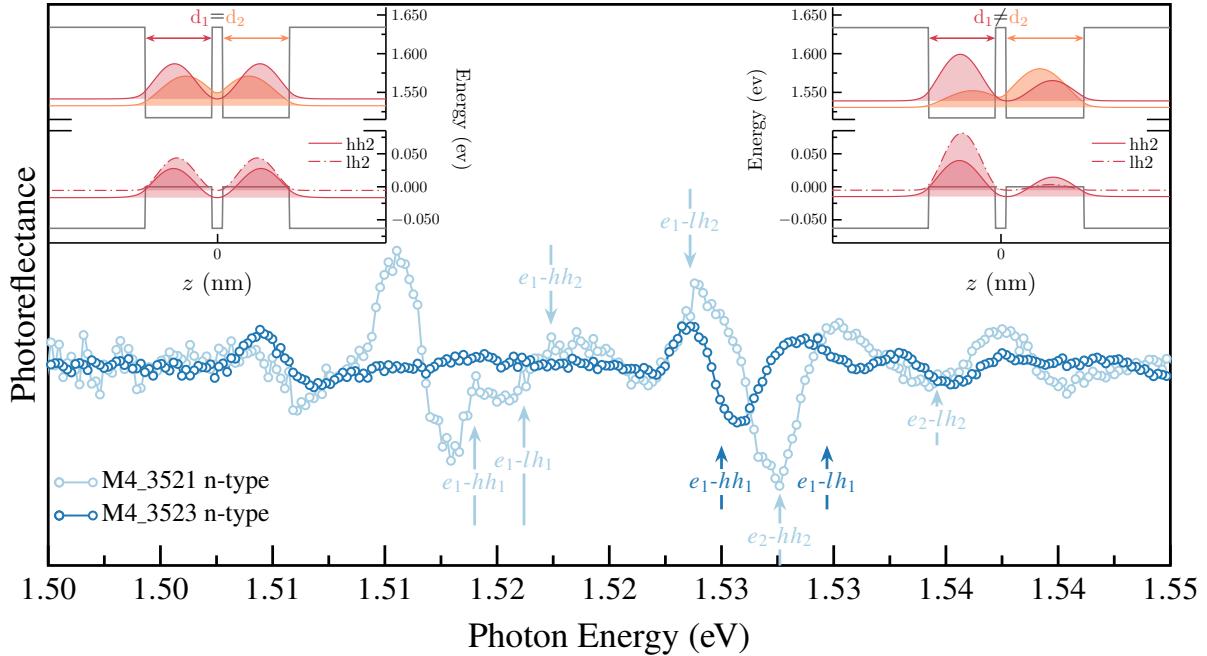


Figure 3.13: The PR comparison between samples M4_3521 (ACQWs-2) and M4_3523 (SCQWs), the electron wave function are plotted for each sample where, the SCQWs sample at top left and at top right to ACQWs sample. The top arrows pointed to forbidden transitions in ASQWs-1 sample, while the bottom arrows pointed to the direct transitions in both samples.

the PR experiments at higher laser power, the results were peculiar due to appearing a higher peak with respect to direct transitions, in fact, the direct transitions didn't was observable. These experiments were realized several times and the behavior was kept, then we decided to increase the spectral resolution to try to understand the nature of this peak, previously all experiments were carried out with the monochromator slits at 1500μ this to enhance the light collected by PM. The Figure 3.14 shows the evolution of these experiments at $P=50\text{mW}$ as a function of the slits aperture, it's clearly that doesn't about of experimental contraption or another external thing which can contribute or being the cause of this behavior.

From these results it decided all experiments with these apertures of slits, although we try to enhance the spectra resolutions at $750\mu\text{m}$ was they got the best results. In the Figure 3.15 shows the evolution of the PR spectra as a function of laser power, starting with 1mW of power and finished with 50mW . Remember previously mentioned that the laser power is unstable and this trouble it didn't allow performing the experiments increases power in one way more uniform, this is the reason that the experiments were carried out with powers at 1mW , 3mW , 8mW , 30mW , and 50mW . The spectra with power less than 8mW are relatively similar, therefore, after that power it can see a change in peak width and that peak tends to shift at less energy.

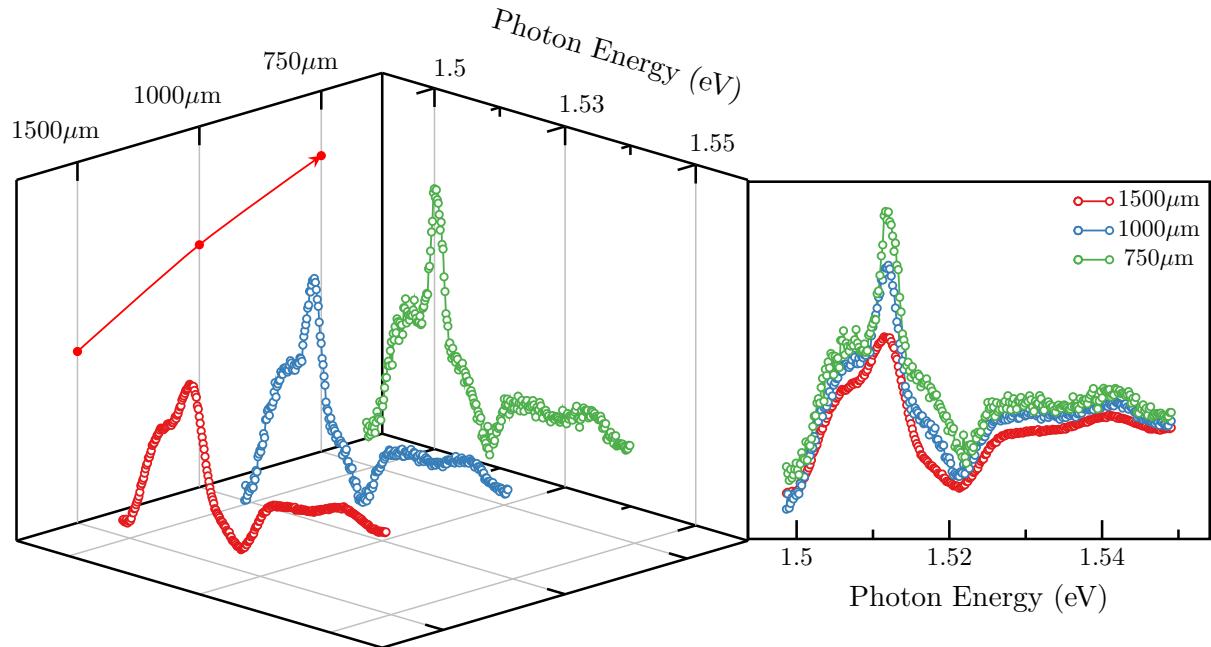


Figure 3.14: PR spectra of the ACQWs-1 sample designed as a function of slits aperture, where it can see the increase of peak resolution as decrease the aperture of slits.

Now, we establish the physical background of that rare behavior in the PR experiments. In semiconductors, the electron-hole pairs are the reason for many special phenomena and are commonly called the hydrogen atoms in semiconductors. But to understand the nature of excitons and their consequent behaviors, it needs to start with a specific platform to keep or extend their life and interactions. As above-mentioned, the life of the excitons in QWs is extended for several reasons, of which; well width and low temperatures enhance the binding energy [159, 160]. When the light interacts with these structures, the electron-hole pair associated with the absorption of a photon with enough energy results in an exciton (general explanation), but if inside the structure the electron density is great [161], the excitons and all possible interactions which can occur as exciton-exciton, exciton-hole (X^-), exciton-electron(X^+), electron-electron, even, LO-phonon-exciton interaction. All of this presents in a modification of the line shape resultant, in terms of the PL experiments it's possible to observe unexpected transitions as a slight modification in the line shape. In spite of the physics involved in this mechanism is very complex, the hard work in this theme has generated valuable results.

Since the 50s Lampert [162] suggested the existence of charged excitons also called *trions* in semiconductors and after almost 40 years it's proved experimentally [161, 163], and as expected the trions X^+ or X^- they were observed in QWs. The electron concentration has a role important in trion formation, for this reason, they usually modulated n- or p-type doping in the QWs structures. Also, the external perturbations as the electric* or

*In this case the trions formation is due a relative position of the Fermi level when the low electric

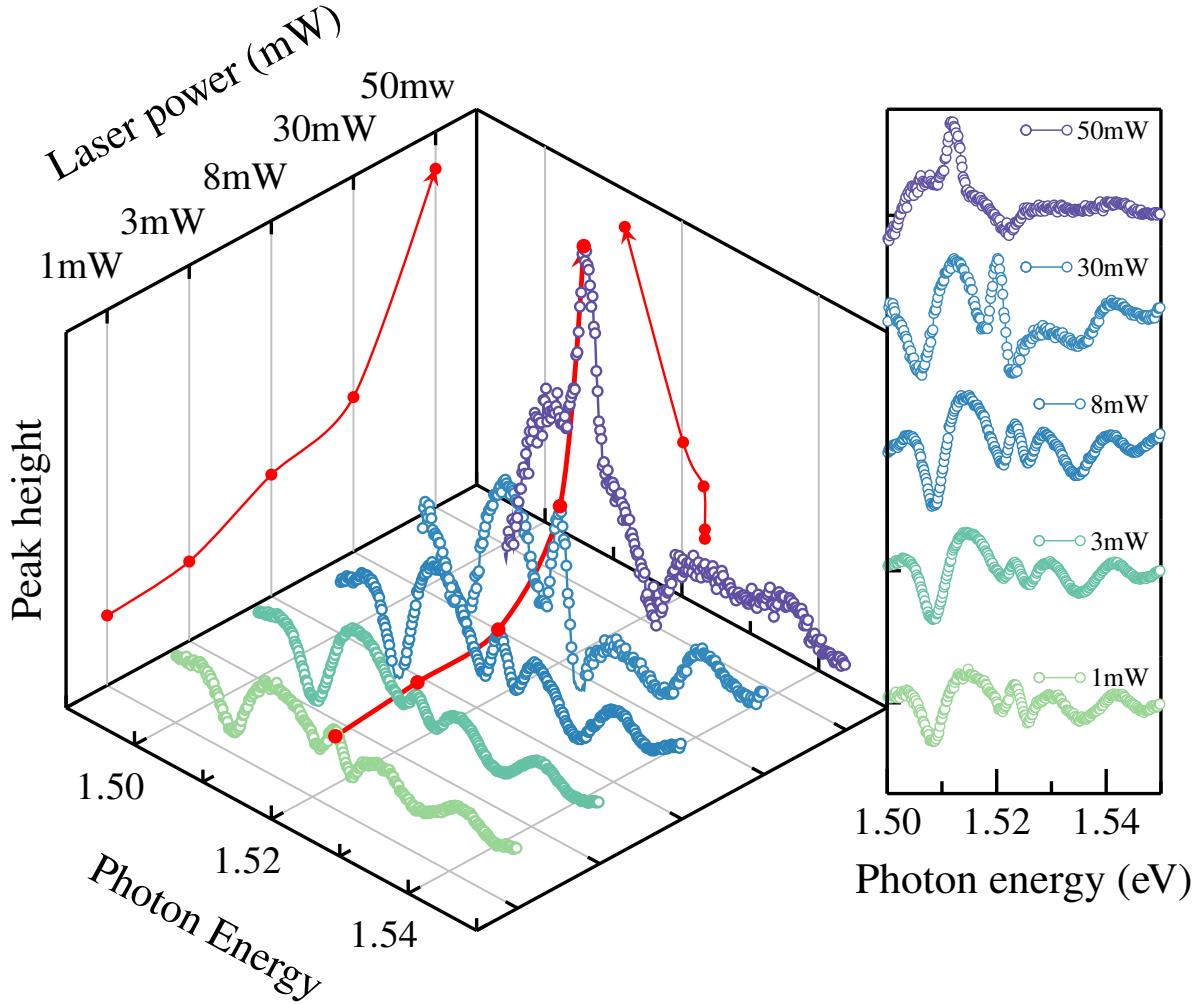


Figure 3.15: The peak tends to height as increase the laser power, which also is very observable as a redshift.

magnetic fields commonly used to enhance the trions transitions, in the magnetic field case, the trions involved acquiring a triplet state nature, so the Zemman splitting is expected, therefore the transitions are well resolved. At zero field, the ground state of trion is a singlet [164], this is, two electrons with opposite spin, these electrons are bound with a hole as shows the scheme in Figure 3.16(a).

In our case, the trions formation is due to photo-excited carriers induced by the laser source, if we suppose that the doped layer is the absolute answer of this, the other samples as SCQWS-1 and ACQWS-2 would have similar results in their PR spectra when it increases laser power, but this is not observed, in fact, in these cases the line shape kept but noisily,

field is applied, in fact, the mechanism involves indirect transitions and is easy that the indirect exciton interact with elctrones or holes [129].

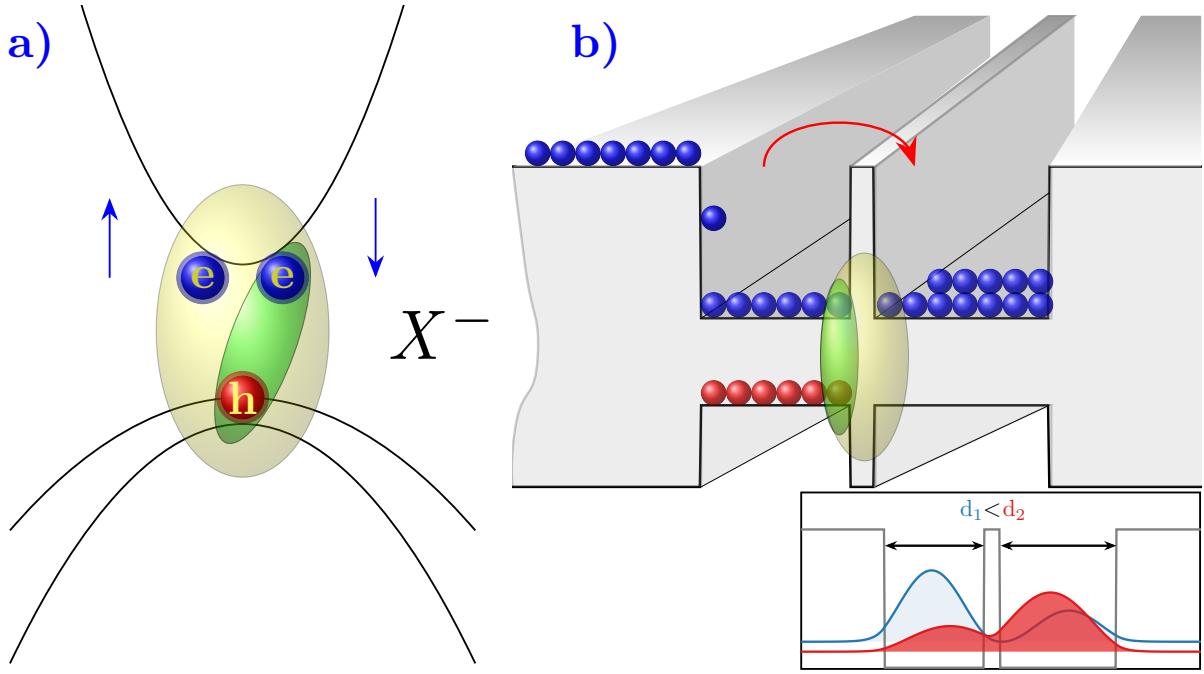


Figure 3.16: Trions formation scheme in terms of band structure (a) in this case, the exciton is bound to an electron in the conduction band leading to a three-body system knew as negative trion \mathbf{X}^- . On the other hand, in (b) it presents the possible formation of \mathbf{X}^- due to a slight width of one in the CQWS, consequently, the narrow well transfers electrons through tunneling to a wide well, if it's calculated the wave functions of this structure, the wave functions have the characteristic of to be distributed asymmetrically as in the case of the applied electric field along z [26, 27].

then if we adjudicate the trion formation to photo-excited carriers, which is the cause that enables trions in the ACQWs-2 sample?. The answer is a hypothesis that needs extra experiments but is part of the future objectives of this work. The electron and holes (hh , lh) wave functions in samples slightly asymmetric in wells widths, i.e., one of the wells is slightly width than the other, these wave functions have similar behavior as the case when de electric filed is applied. This behavior enhances the electron tunneling from the narrow well to the other well (wide well) as shows in Figure 3.16(b), if all-important samples contain the doped layer with the same n-type concentration if we suppose that this layer modifies the Fermi level thus generating a small electron gas (2DEG), its possible think that this electron gas has a function of electron reservoir [165, 166]. Then the photo-carriers generates by the laser source step up carriers dynamics, doing that the narrow well is yielding continuously electrons to wide well through tunneling and, these electrons are recombining with the excitons confined in the plane of the narrow well, therefore results in a three-body system $\mathbf{X}^- = (e, e, h)$ or $\mathbf{X}^+ = (e, h, h)$. As it's known, the trion is a charged exciton where the sign depends on its formation, in the case of $\mathbf{X}^- = (e, e, h)$ their transition is under the first transition of X_{hh} , and their energy evolution tends to redshift as can see in Figure 3.15. Therefore in our case, which was

also has been reported [161, 165–168], the shift evolution correspond to an \mathbf{X}^- trion, however still missing more experiments to strengthen this hypothesis. On the other hand, what happens if the 2DEG doesn't consider? It is very important to emphasize this argument because the objective of this work is to demonstrate that CQWS structures especially ACQWs shows effects of symmetry breaking doesn't see in structures without external perturbation(application of: electric or magnetic field, strain, etc.) or intentional modification (growing of interfaces that unbalance the QWs region, differences in the potential of the barriers), by this reason is importantly empathize that regardless of exist a 2DEG whichever is their electron density and the built-in electric field which can this generate, as a matter of fact, that field doesn't represent the cause of the phenomena presented in this work, more later this is discussed with more detail. On the other hand, is relatively easy to corroborate that the presence of the electric field on those samples can be regarded as despise, what is the reason to asseverate this if the PR has a principal characteristic the built-in field modulation? As before mentioned the non-existence of the FKOs is a point to assert the field regime is low, moreover, in comparison with the n-i-n-type samples, also remain in this regime notwithstanding of be designed to reduce the built-in field. However, it's can be estimated by means of Schrödiner-Poisson, so as to, the equation Equation (2.5) it's coupled Poisson equation [24, 169]

$$\left(\frac{d}{dz} \varepsilon(z) \frac{d}{dz} \right) V_p(z) = \rho(z) \quad (3.7)$$

$$\left(\frac{d}{dz} \varepsilon(z) \frac{d}{dz} \right) V_p(z) = e \left[n_D(z) - \sum_i n_i^s |\psi_i(z)|^2 \right] \quad (3.8)$$

With the objective of present the behavior and the causes of the high doping in the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layer, we implement a simple code, starting off our numerical codes and helping us with already implemented codes as Aestimo [7], we calculated numerically Schrodinger-Poisson equation. It is important to mention which solution is self-consistent, therefore the code is implemented with all parameters to divergence avoid, in our case is due to high doping and this is too large.

For this reason, it is inevitable that the codes don't converge, although it can be considered a factor damping to speed convergence [170]. We consider the damping factor, and we decided to calculate a structure as GaAs/n-type doped $\text{Al}_x\text{Ga}_{1-x}\text{As} / \text{Al}_x\text{Ga}_{1-x}\text{As}$, where the width of lateral layers is fixed and the width of the doped layer varies from 15nm to 300 nm with the same n-type doped $6 \times 10^{18} \text{ cm}^{-3}$. In general, the self-consistent Schrodinger-Poisson equation, is a process that starts with the calculation of the confined energies in the potential profile defined as $V(z)$, in this profile are included each parameter of the material that makes up the heterojunction as the doping quantity in each layer if this is doped. After, as shown in Equation (3.8), is evaluating the space charge with their respective charged donors and their concentration n_D , n_i^s is the electron sheet density of the confined levels and corresponding wavefunctions $\psi_i(z)$. To calculates the electron density in each level i frequently is applied Fermi-Dirac statistics [43, 171, 172].

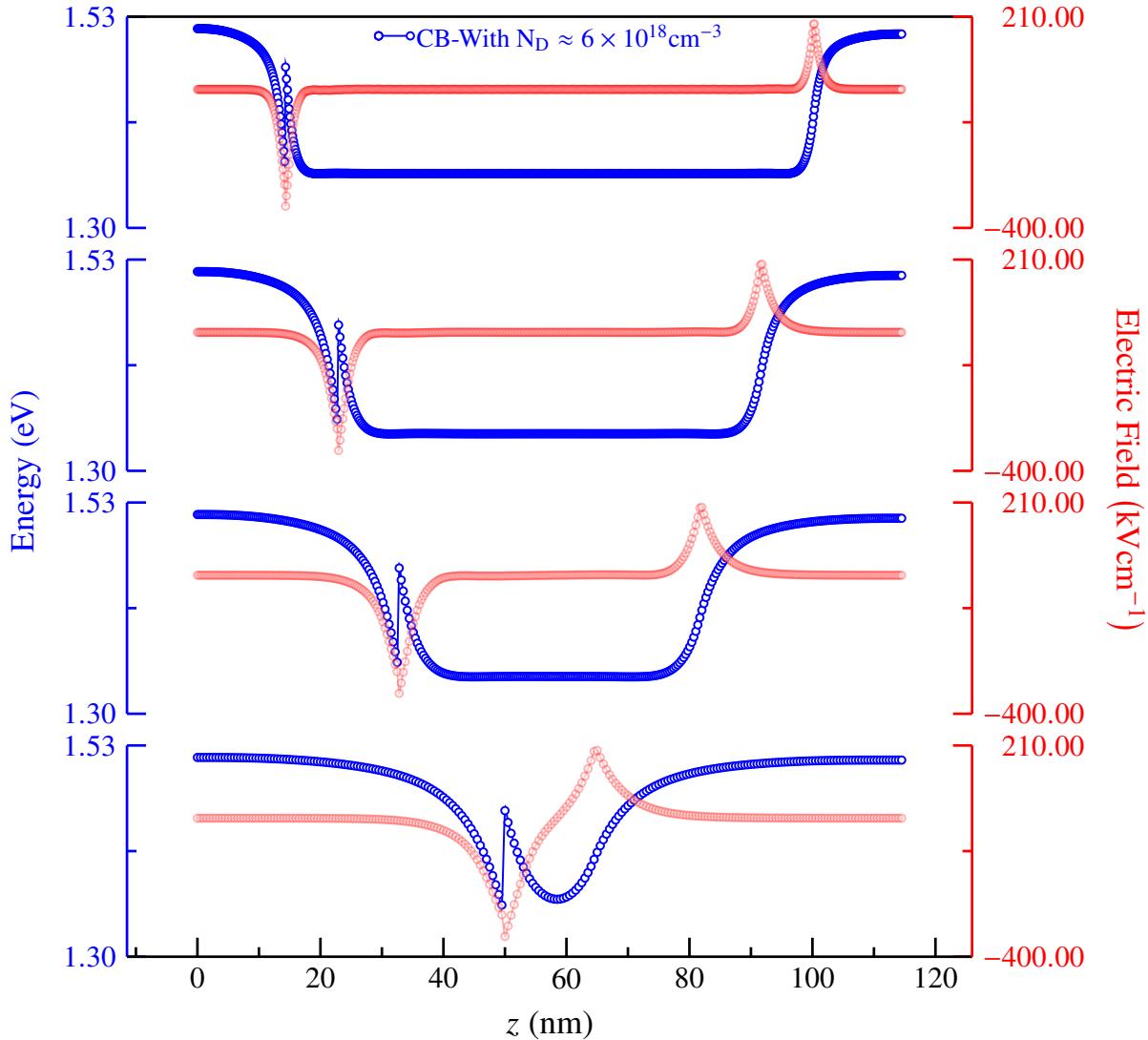


Figure 3.17: Results to self-consistently Schrodinger-Poisson equation, in order with down to top the n-type layer $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ doped ($6 \times 10^{18} \text{ cm}^{-3}$) is increasing in width from 15nm, 75nm, 150nm to 300nm. The goal of this is to calculate in a general way the effects of the doped layer, specifically due to the electric field induced by this. The strength of the field is expressed in kVcm units, although this magnitude si great, we assume that doesn't significantly, latter we explained this.

This charge distribution in the structure gives rise to space charge effects, resulting in an additional electrostatic potential V_p which causes conduction band bending [169, 173]. The total potential V is the result of $V = V_0 + V_p$, where the V_0 is the original potential profile, so, this is the iterative part of calculations, in our case, we established the difference between $e_{1\text{new}} - e_{1\text{old}} < 1 \times 10^{-5}$ as convergence factor. Previously mentioned the damping factor is defined for fast convergence, in our case α_{damp} is about 1×10^{-3} . The results are shown in Figure 3.17 to a structure : $\text{GaAs}/\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}(\text{n-type } 6 \times 10^{18} \text{ cm}^{-3})/\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ with four different widths (15nm, 75nm, 150nm, and 300nm) for the layer doped.

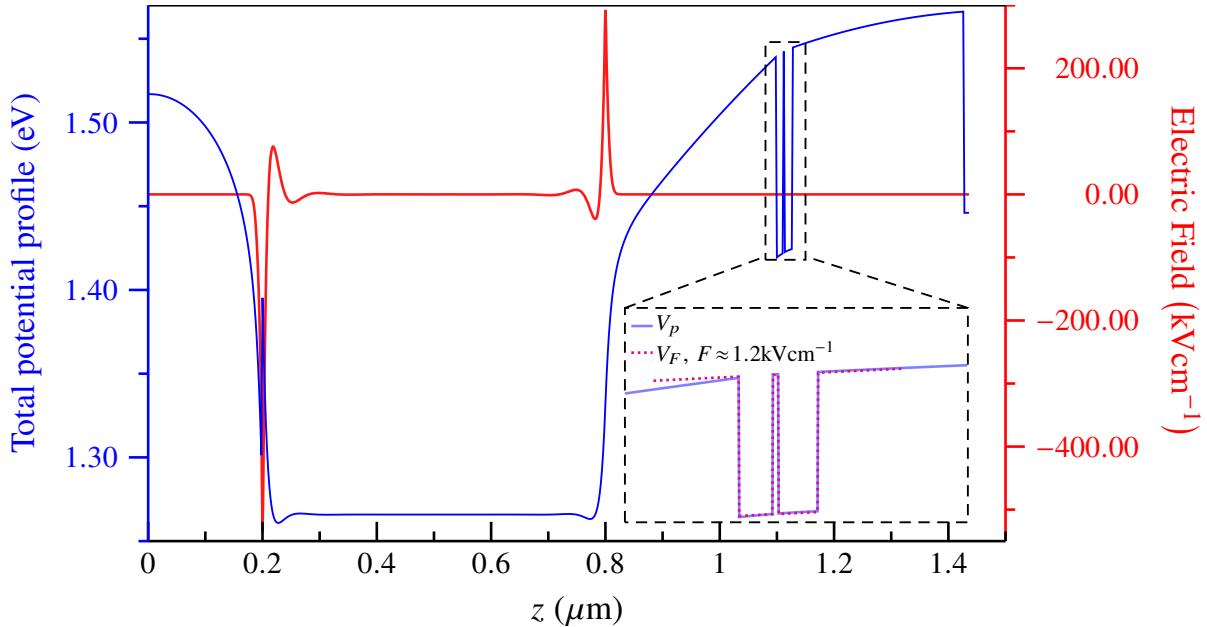


Figure 3.18: Band conduction profile $V(z)$ calculated by numerical solution of self-consistent Schrödinger-Poisson equation. The calculations were performed considering the width of the doped n-type 6×10^{18} layer with 600nm. The zoom inset shows the comparison between total potential calculated (blue) and when applied field $F \approx 1.2\text{kVcm}^{-1}$ (dotted magenta), where at around of CQWs zone are similar.

In the next chapter, we expose the RAS experiments and their significance in the wonderful results of this work, following, is important to define and explain the role of the doping layer that as can see may be generated a significant intrinsic field which in contrast with the RAS results and our model the built-in electric field doesn't significatively important in the symmetry breaking ($D_2 \rightarrow C_{2v}$). Therefore, we calculate the possible conduction band bending due to the built-in electric field as can see in Figure 3.18.

We have taken into account a total structure width with an n-type doped layer, we remember that the high doping and large width can cause the calculations doesn't converge by this we implemented a damping factor to accelerate the convergence, also we calculated a potential profile considering an external electric field applied around of $F \approx 1.2\text{kVcm}^{-1}$. The results show that practically the total V_p and electric field (line-shape and magnitude) are kept as shown in Figure 3.17, if we compare the potential profiles V_p and V_F as see in Figure 3.18, it can observe that practically the band bending which is generated by electric field so much as by doped layer, as an external field applied are very similar if the external potential is around of $F \approx 1.2\text{kVcm}^{-1}$. This means that, if exists an electric field but is comparable with surface field [174], hence is a small field. Therefore, we can say that the effects of the trions are associated with the asymmetry of the QWs as before mentioned.

3.2.2.2 The PR summary

In conclusion, the PR remains a powerful tool for experimental solid-state physics, especially in semiconductors study the facility to implement it, and the great information which gives as about fundamental transitions that in comparison with other spectroscopy is still better. Along with the experimental work, we could notice that the PR has the capability of detect behaviors which doesn't common in this spectroscopy as trions measured, and although this work is still in progress, the satisfaction to propound a novel source to study of an excitonic behavior as the trions, through easy spectroscopy without external perturbations. On the other side, the AQCWs has a large potential to study quantum phenomena, especially the interactions and process due to the exciton confined, in this case, something so simple as the relative widths in the CQWs generates a surprising behavior.

3.2.3 Reflectance Anisotropy Spectroscopy (RAS)

THE RAS is the experimental tool that completes the set in this work, without the intention of replicates the physical background and interpretations about RAS, we focus on specific terms to detail our great results. This spectroscopy, is a powerful tool in the studying of semiconductors physics, being characterized as default anisotropy study tool. This experimental technique, was developed by Aspnes [145, 175, 176] to measure *surface-induced* optical anisotropy in cubic semiconductors, although this can be applied around of near-band-edge [177]. So, to our purposes, RAS is an excellent experimental tool to study optical anisotropies in CQWs structures. In our case both RAS and PR setup is the same with their exceptions, in the PR case is necessary to add the laser to modulated spectroscopy while in the RAS the modulation it's realized by the PEM, which changes the polarization state. As schemes in Figure 3.19, the monochromatic light first times through over polarizer prism and the PEM to finally being focused onto the sample with spot size of 5.00mm diameter. The light reflected by the sample is collected and detected by the multialkali photomultiplier tube (before discussed in Section 3.1 and shows in Table 3.2). A detailed description of the RAS technique can be found elsewhere [178]. As shows in Figure 3.19, the RAS signal is proportional to:

$$\frac{\Delta R}{R} = 2 \frac{R_{[110]} - R_{[1\bar{1}0]}}{R_{[110]} + R_{[1\bar{1}0]}} \quad (3.9)$$

where denotes the orientations $[110]$ and $[1\bar{1}0]$ over crystalline directions(see Figure 3.1). In our experiments the RAS signal is around of 10^{-4} . As well's known if the RAS signal it's detected, the structure exhibits an optical anisotropy for in-plane light propagation [12]. The experiments, as the PR case, were it performed at 30K.

In accordance with Chapter 2, the anisotropy into these structures entails into interesting physical phenomena, overall about of optical properties. This IOA it's due to the hole mixing, as a long as the structures being under symmetry reduction, in this case from $D_{2d} \rightarrow C_{2v}$. If we measured RAS over SCQWs, it's expected that this doesn't exhibit an IOA or failing that this being smaller due to abrupt interfaces are non-ideal.

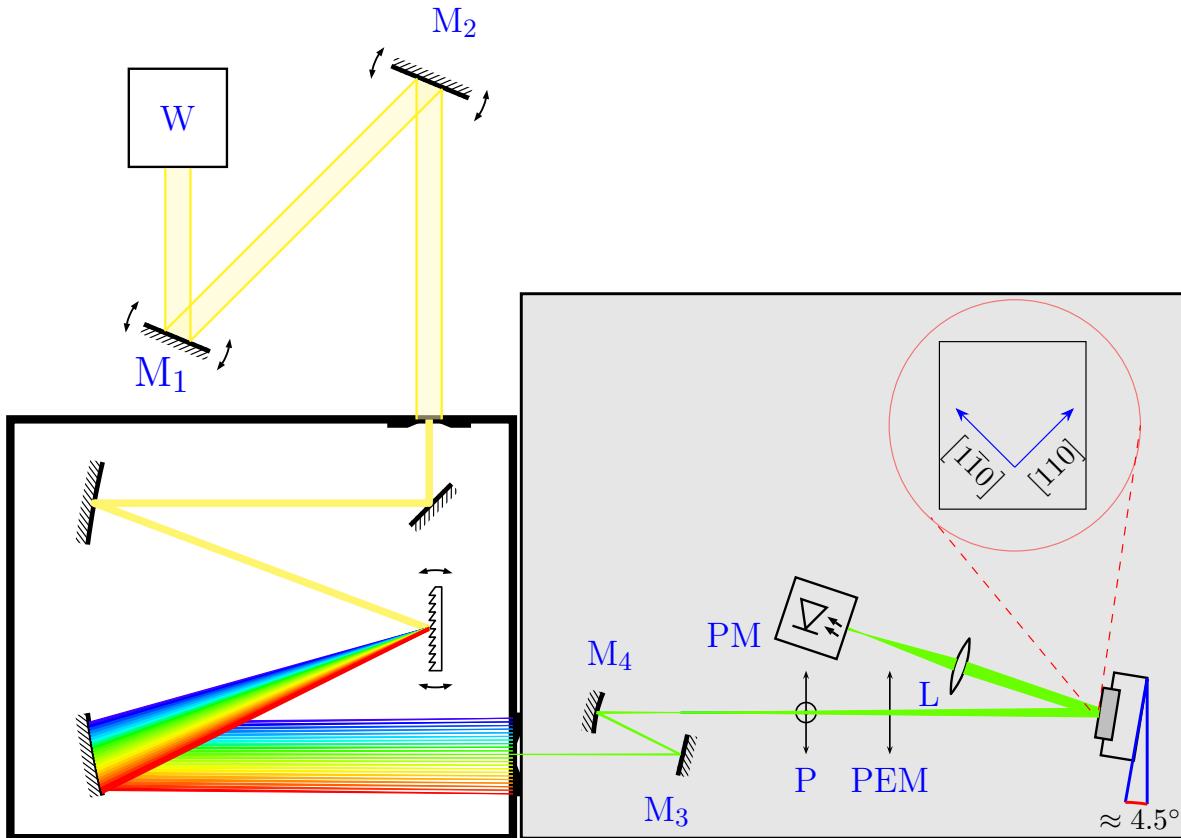


Figure 3.19: RAS setup implemented in this work, as before explained in the PR setup, this is a dark configuration this due that photo-detector are exposed, then keeping closed to ambient light. The optical array is the same as the PR, with the difference which the role of polarization and PEM. This figure also schemes the incidence angle which is about of 4.5° and the directions of linear polarization.

The first part of experiments were carried out over the samples: M4_3171, M4_3172 and M4_3226, we remember that these samples are the type n-i-n, this means that they consist in more growth layers, therefore it's expected that the RAS signal being smaller. The Figure 3.20 shows the results to these samples, in top to bottom order, in left side plots the RAS while in right side it's plotted the R spectra. The R spectra plotted to each sample is the average of all experiments performed, this means which in each RAS experiment, the glsR was simultaneously measured, then to each sample it's taking the average. In order to discuss the result of that samples, we can observe that in general thermally these samples gets a smaller signal of RAS, although in R it does not happen, the direct transitions can

being locate in accordance with the numerical results as shows with arrows.

Although these three samples consists in more several growth layers, it's possible to measure an in-plane anisotropy were in samples: M4_3171 and M4_3226 the signal amplitude it's relatively same in contrast with the sample M4_3171.

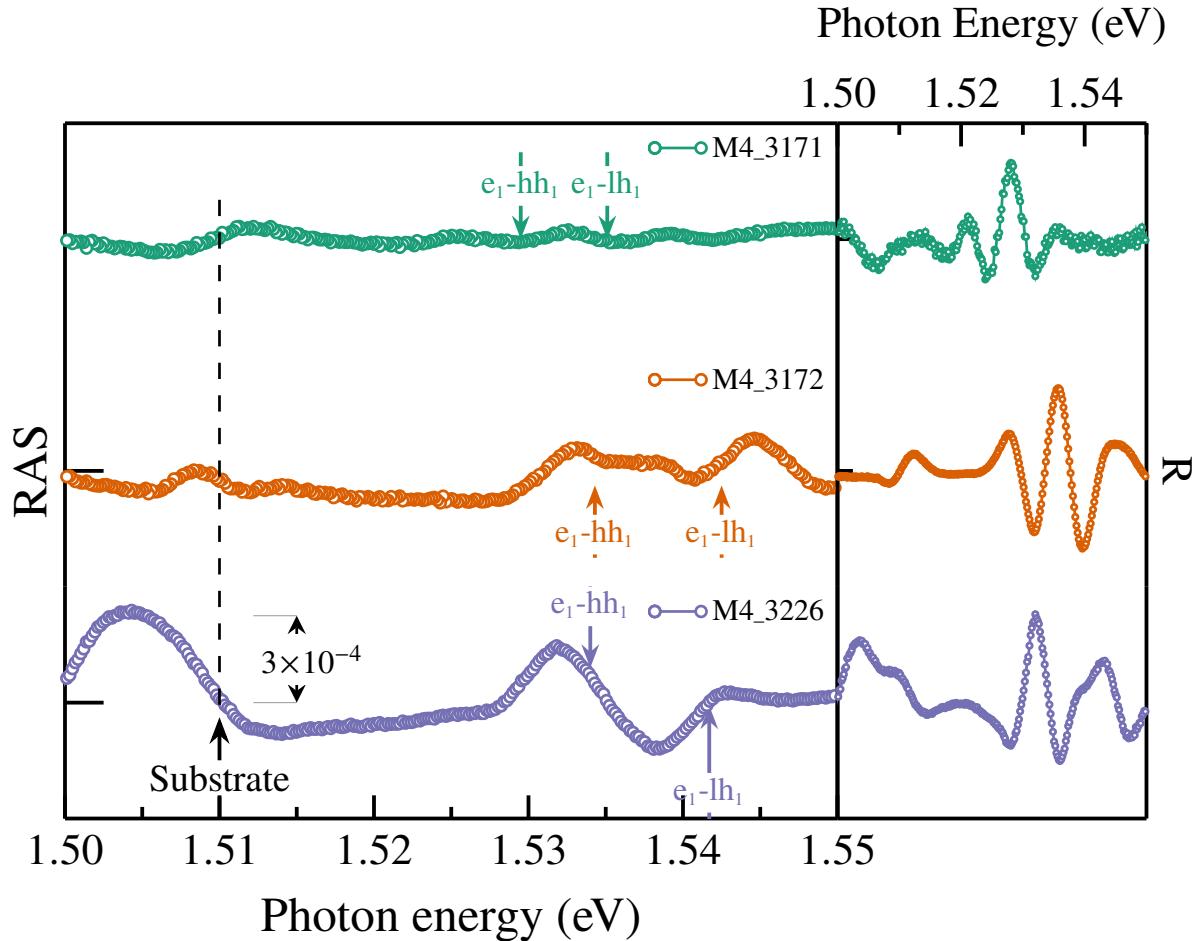


Figure 3.20: Experimental results from samples: M4_3171, M4_3172 and M4_3226, from top to bottom respectively. In left, shows the RAS result where with a dashed line it's denote the substrate transition, in each sample denotes the direct transitions with arrows. The right side, show the plots of R spectra, which is the average result of all experiments carried in each correspond sample.

The first RAS results even they are an asymmetric CQWs structures, the RAS spectra it's higher in the samples with AlAs barrier (M4_3172 and M4_3226), then the potential barrier is higher than in the sample M4_3171, but the width of these barriers is small, therefore we have a case with samples they consist; in high coupled barrier and thinner. In the other sample, where the RAS spectra is smaller (M4_3171), the coupled barrier is small in potential in comparison with the other samples, but is more wide.

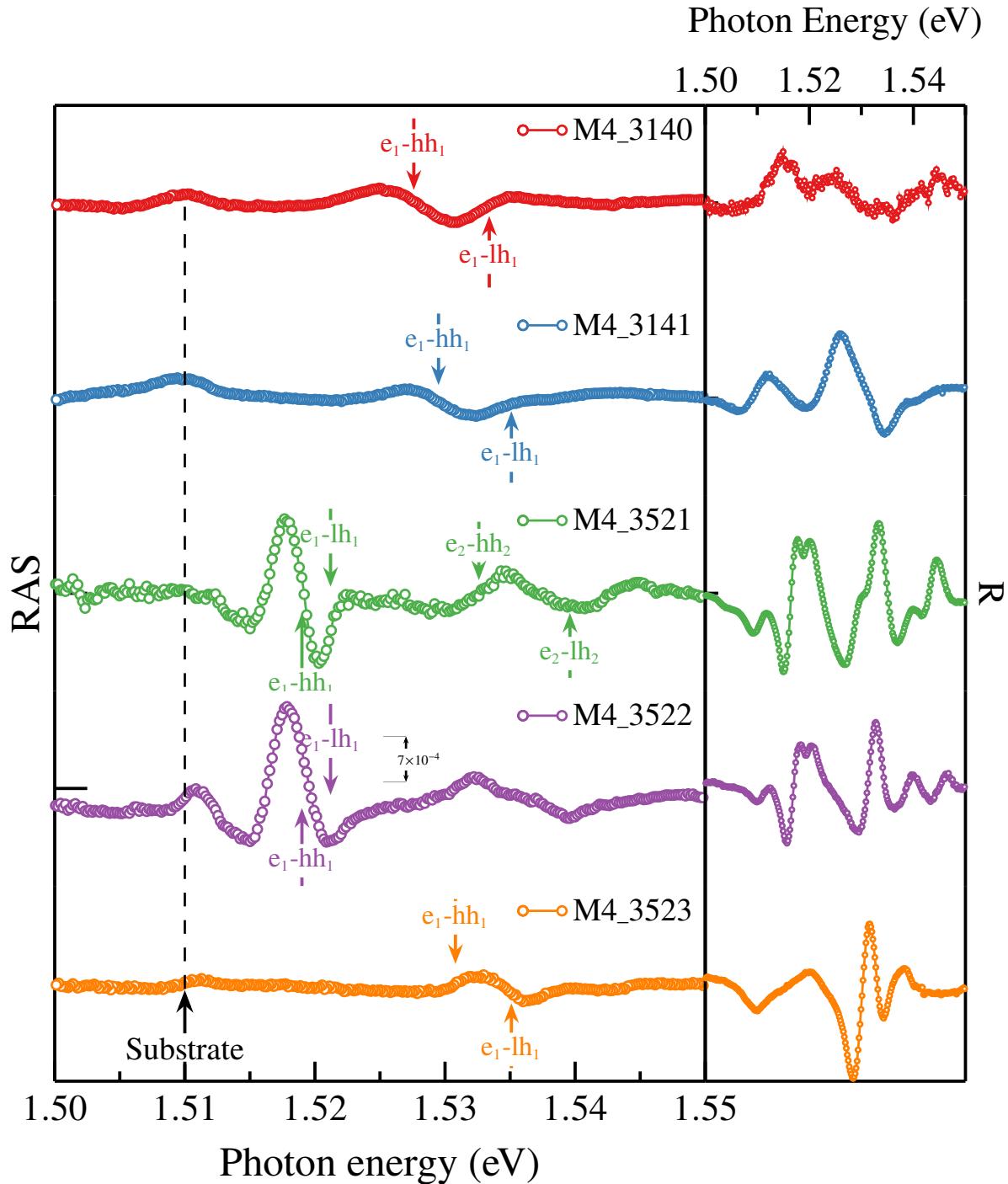


Figure 3.21: Experimental results from samples: M4_3140, M4_3141, M4_3521, M4_3522 and M4_3523, from top to bottom respectively. In left, shows the RAS result where with a dashed line it's denote the substrate transition, in each sample remarks the direct transitions with arrows. The right side, show the plots of the R spectra, which is the average result of all experiments carried in each correspond sample. Also, it's denote the RAS magnitude proportional to 7×10^{-4} , which in contrast with SCQWs samples the signal is smaller. The direct transitions it's locate by two peaks with opposite concavity where can see that the larger one transition is $e_1\text{-}hh_1$ and smaller one associated with the $e_1\text{-}lh_1$.

Although, it was expected that the spectra relatively same in these three samples, the coupled barrier plays an important role this due to the tunneling process, wells it returns a RAS spectra more definite (better line-shape) than the sample with less tunneling, this means, the coupled barrier is wide. Then, the sample M4_3171 is approx three times smaller. These samples are relatively common with barrier exception, in sample M4_3226 that better has RAS response posses a coupled barrier with a width of 0.424 nm, while the second with a better RAS response is the sample M4_3172 which has a barrier width of 0.565 nm.

In the next set of samples: M4_3140, M4_3141, M4_3521, M4_3522 and M4_3523, as is explained in Section 3.1, consists in samples with coupled barrier of $\text{Al}_{0.15}\text{Ga}_{0.9}\text{As}$, where the difference in this set samples, is one of these samples (M4_43141) has a coupled barrier twice wider than the other samples, and the relative width of the wells.

The Figure 3.21 shows the results of RAS experiments carried out on these samples. These results are interesting in comparison with the before set, the most notorious is the evolution of RAS signal in the more asymmetric structures, in these structures the peaks associated with direct transitions are seen more clearly, being those with an opposite concavity. In these peaks the larger one it's the heavy-hole (down concavity) while the smaller one it's the light-hole transition (up concavity). It's totally evident that the samples M4_3521 and M4_3522 (also called as ACQWs-1 and ACQWs-2) exhibits great RAS response, then, in accordance with the model anisotropy has a major hole mixing. Also, the R spectra is well resolve in both samples, with a peculiarity in the transitions concavities, which in both, is up concavity.

3.2.3.1 RAS strength discussion and the physical model justification

To enter into discussion, its purpose is to focus on the three samples before mentioned, aiming to expose the principal objective of this work, which is denoted the RAS strength in ACQWs as importance in the optical properties and excitonic effects in these structures. The Figure 3.22 exposes the evolution of RAS strength, in this plot it is indisputable that the signal increases, because the comparison between the three samples where it's starting with a symmetric structure, then with a lightly asymmetric structure and finally with a very asymmetric structure, the RAS signal associated with each transition have opposite concavities and redshift as the structure is more asymmetric.

Due to the fact that the wave-function probability density of sample SCQWs is distributed symmetrically along the QW structure, the IOA strength is expected to be similar to that obtained for a single QW. In fact, a RAS signal of the order of magnitude of 0.1×10^{-3} has been reported for an 8 nm single QW [75], which has the same order of magnitude as the spectra of Figure 3.23(c). This IOA is attributed to the inequivalent AlGaAs/GaAs interfaces along the SCQWs structure. It's important every clear the role of tunneling in the coupled wells, the difficulty to get a model to explain this structures doesn't it the

same as the single QW structure.

As pointed out before (Section 2.5), the strength of the IOA signal is produced by an intermixing of the heavy- and light-hole states in the valence band that is proportional to $\langle \psi_{hhn} | \mathcal{H} | \psi_{lhn} \rangle$ according to Equation (2.13). For the lowest heavy- and light-hole levels ($n = 1$), there is an estimated separation in energy of around $\Delta E_1 = 2.0, 4.1$, and 4.4 meV for samples ACQWs-2 (ACQWs-3), ACQWs-1, and SCQWs, respectively. The mixing $\langle \psi_{hhn} | \mathcal{H} | \psi_{lhn} \rangle$ can be estimated by considering that the transitions are direct (n is the same for the valence and conduction band) and then the overlapping terms in Equation (2.13) must be approximately the same for each sample. For transitions $n = 1$ it can be seen in Figure 3.22 that amplifier ratios of the spectra between ACQWs-2 and ACQWs-1 with respect to SCQWs are 1.5 and 3.7, respectively. Thus from Equation (2.13) we estimate ratios of $\langle \psi_{hh1} | \mathcal{H}_{ACQWs-1} | \psi_{lh1} \rangle / \langle \psi_{hh1} | \mathcal{H}_{SCQWs} | \psi_{lh1} \rangle \sim 1.4$ for sample ACQWs-1 and $\langle \psi_{hh1} | \mathcal{H}_{ACQWs-2} | \psi_{lh1} \rangle / \langle \psi_{hh1} | \mathcal{H}_{SCQWs} | \psi_{lh1} \rangle \sim 1.7$ for sample ACQWs-2.

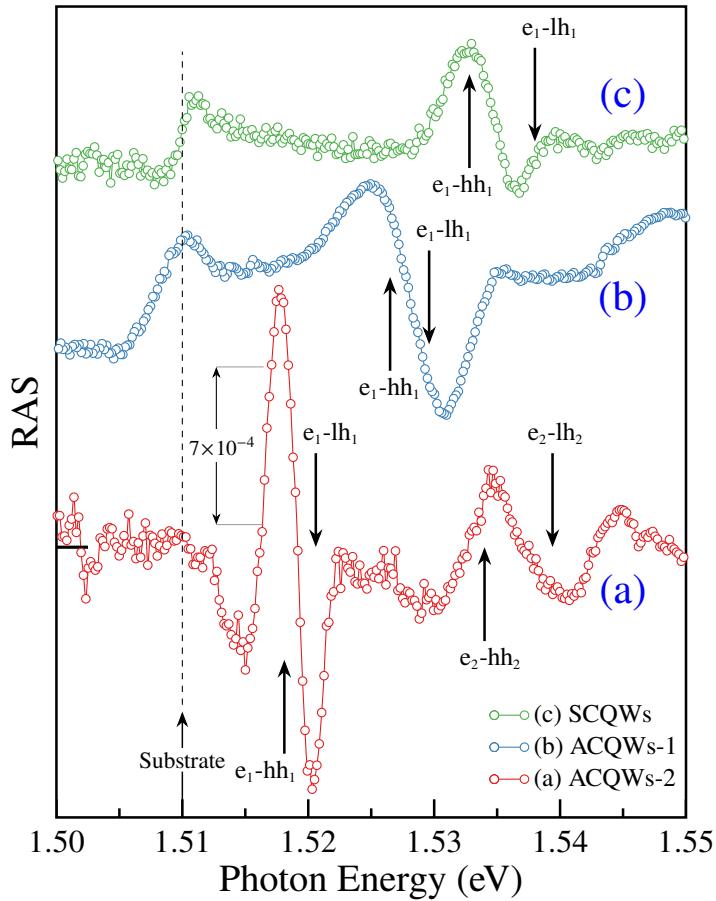


Figure 3.22: RAS spectra for the (a), (b), asymmetric and (c) symmetric CQWs. The dashed vertical line indicates the expected energy of the excitonic transition of the GaAs substrate. Above this energy, the optical transitions come from the DQWs. The inset shows the PL spectra measured for each sample. Two peaks can be identified in each spectrum, a larger one associated with the transition e_1-hh_1 and a much smaller one associated with the e_1-lh_1 [for spectrum (b) this peak is observed as a shoulder]. The energies obtained from the PL spectra are indicated by the arrows in the RAS spectra. Note that the structures associated with e_1-hh_1 and e_1-lh_1 increase their strength when the DWQs become more asymmetric. The RAS spectra were measured at 30 K.

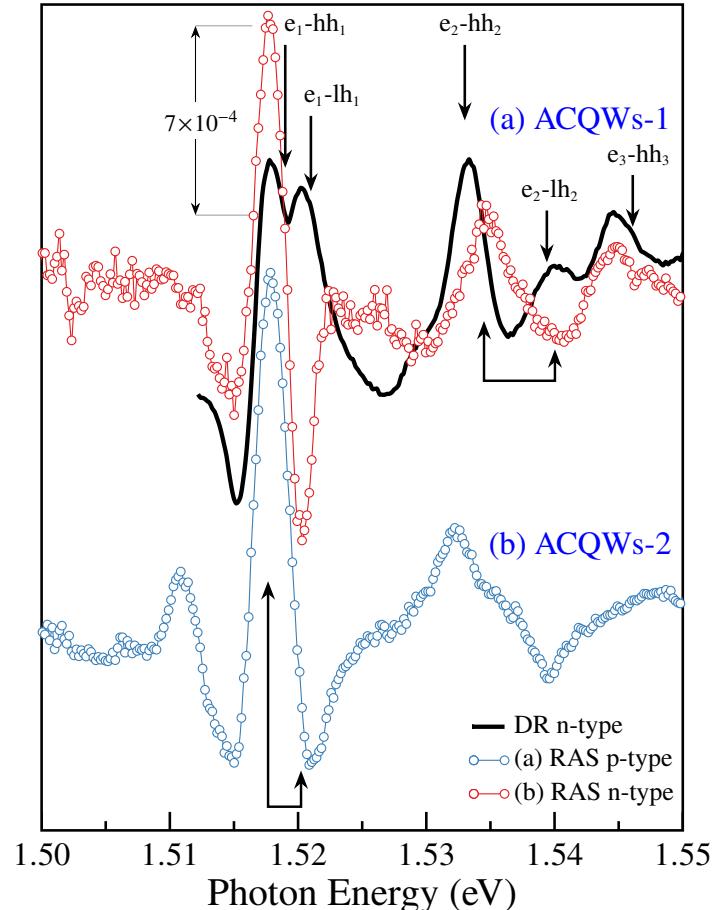
In accordance with our estimation based on the anisotropy model, the Table 3.5 contains the energies considered. If well, the large IOA in general, it's attributed to interfaces in our case the reason is due to non-equivalence due to the width asymmetric into coupled QWs, this originates a mixing of hh-lh states and therefore these it coupled strongest as the

Sample	e1-hh1	e1-lh1	ΔE_e (meV)	ΔE_{hh} (meV)	ΔE_{lh} (meV)	ΔE_n (meV)
SCQWs	(N)1.5328 (E)1.5297	(N)1.5380 (E)1.5341	8.9	0.9	6.9	$\Delta E_1 = 4.4$ $\Delta E_2 = 10.5$
ACQWs-1	(N)1.5265 (E)1.5273	(N)1.5296 (E)1.5314	8.4	1.3	6.4	$\Delta E_1 = 4.1$ $\Delta E_2 = 9.2$
ACQWs-2	(N)1.5181 (E)1.5190	(N)1.5206 (E)1.5210	11.8	3.4	7.5	$\Delta E_1 = 2.0$ $\Delta E_2 = 6.4$

Table 3.5: Comparative of experimental (E) and numerical calculations (N) of first level transition energies (in eV). δE_e , δE_{hh} and δE_{lh} corresponds to the difference between electrons, heavy- light holes states, respectively. ΔE_n is the numerical calculation of energy splitting for transitions 1 and 2 ($n = 1, 2$).

states close in energy [81]. The Table 3.5 and Figure 2.10 confirms this, in the ACQWs-2 this energy between hh1 and lh1 states are close, in counterpart with the SCQWs this energy is large.

Figure 3.24: Reflection anisotropy (RAS, (a), (b)) and differential reflection (DR, solid line) spectra for ACCQWs-2 and ACQWs-3, grown on an AlGaAs n-type and p-type layer respectively. Note that while for the heavy hole transitions ($e_1\text{-}hh_1$ and $e_2\text{-}hh_2$) in the RAS and DR spectra have the same concavity, for light holes transitions ($e_1\text{-}lh_1$ and $e_2\text{-}lh_2$) the concavities are opposite and DR spectra shows the highest level transitions. The bottom arrows point to the experimental transitions for the two first levels, whereas the top arrows show the calculated energies to three energy levels. The RAS and DR spectra were measured at 30K.



In order to elucidate the physical origin of the RAS in the ACQWs, we compare in Figure 3.24 the RAS and the DR spectra of the ACQWs-2. DR spectra is obtained by the numerical subtraction of the reflection (R) spectra recorded at 30K and 300K followed by the normalization to the 300K spectrum. The subtraction highlights the excitonic features, which are very weak (and energy shifted) at 300K. The comparison between RAS and DR spectra allows us to contrast the contribution of the heavy and light holes transitions. Around 1.5175 eV, the DR spectrum shows two peaks corresponding to e1-hh1 and e1-lh1 transitions. Note that in the RAS spectrum, the structure associated to e1-hh1 transition has the same concavity as the corresponding for the DR spectrum, while the e1-lh1 transition has the opposite concavity. This is an indication of the transfer of oscillator strength between the levels due to the intermixing of heavy- and light- holes, thus supporting our anisotropy model. The same behavior applies for the e2-hh2 and e2-lh2 transitions at around 1.5375 eV. Transition e2-hh3 is also indicated and it has the same concavity for RAS and DR spectra, as in the case of the e1-hh1 and e2-hh2 transitions. The arrows at the bottom of Figure 3.24 indicate the energy of the states en-hhn and en-lhn (for $n = 1$ and 2) obtained from the maximum and minimum of the RAS spectrum. From the numerical calculation results summarized in Table 3.5, the energy splitting between transitions en-hhn and en-lhn are $\Delta E_1 = 2.0$ meV and $\Delta E_2 = 6.4$ meV. In accordance with Equation (2.13), the IOA amplitude is proportional to $1/\Delta E_n$. Considering the same value for the overlapping and the mixing $\langle \psi_{hhn} | \mathcal{H} | \psi_{lhn} \rangle$ we estimate an amplitude ratio of 3.25 between these transitions. This value is close to the value of 3.9 obtained by the RAS spectrum of Figure 3.24 supporting our interpretation.

Finally, we discuss the possible contribution to the RAS amplitude by an built-in electric field accross the CQWs both symmetric and asymmetric. To study this contribution, we have compared the RAS spectra of asymmetric samples ACQWs-2 and ACQWs-3. The difference between them is the doping of the AlGaAs layer (see Table 3.1). While for ACQWs-2 it is n-type, for ACQWs-3 it is p-type. Assuming that the built-in electric field originates from charge transfer between surface states and the AlGaAs doped layer (n or p), this field is expected to have opposite signs for samples ACQWs-2 and ACQWs-3 . Thus, the linear contribution of the electric field to the RAS should be reversed in sign for such samples. Figure 3.26 shows the comparison between the RAS spectra of samples ACQWs-2 and ACQWs-3, this with aim to demonstrate that in the case of existed a field the sign is opposite. The experiment, it was designed to measure both samples in a sequence way along the same preferential direction, in this case we choose the direction of pits [1̄10] [28], if existed a field we expected a opposite RAS signal, but this doesn't occur, the sign in signal it was conserved in both samples. The Figure 3.26 shows the results of this experiment, in top left and right it's placed the images of both samples of the pits orientation, this to corroborate that the direction of measured. As can be seen (Figure 3.24), RAS spectra are equivalent in shape and have the same sign, thus indicating that the contribution of the electric field to the RAS signal is very small.

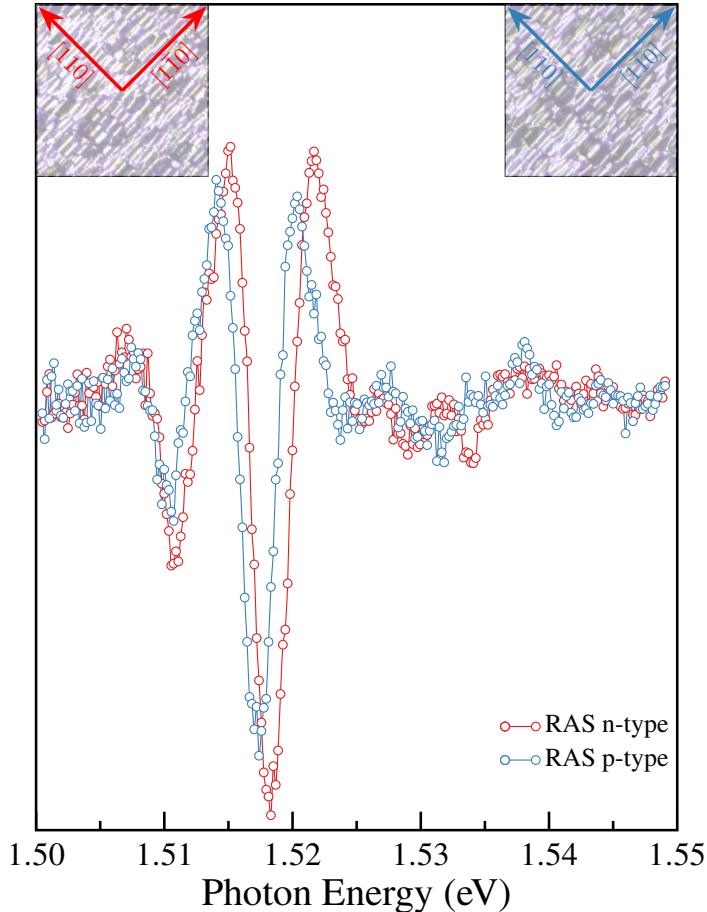


Figure 3.26: RAS experiment designed to demonstrate the non-existence of built-in electric field through sequential measurement along the preferential direction, in this case, it was chosen along the pits $[1\bar{1}0]$ [28]. The signal result in both samples practically is the same, the sign is conserving. At top left and right located the images taken with a microscope of back substrate which shows the pit reveals along of $[1\bar{1}0]$ direction.

It's natural to suppose that the built-in electric field contributes to increase of the RAS signal, and although we estimate that this field is small, it is still important to discuss it and show different ways to affirm that the built-in field isn't the reason of the IOA. We conclude, thus, that the dominant contribution to RAS spectra is the asymmetry of the CQWs system.

3.2.3.2 The RAS summary

In conclusion, the RAS proves experimentally that the IOA increases as a degree of asymmetry in CQWs structures. This behavior is attributed to the reduction of the symmetry of the electronic states in the CQWs with QWs of different thicknesses. The nature of the transitions was identified by using PL spectroscopy and numerical calculations. We believe that the results presented in this work are important and will be useful in understanding the evolution of the optical transition induced by the breakdown of translation symmetry in asymmetric CQWS.

4

CONCLUSIONS AND FUTURE WORK

In this chapter raise the principal conclusion of this work and present the future experiments on these structures in the wake of obtained results.

THE aim of this work finally results in important publications, the CQWs continue to be an excellent platform to study optical and quantum-mechanical properties, this work presents an important result and enhance the importance of study these structures. The principal idea to exposes our results was planned to simplify but specify the physical basis, starting from explain a single QW and structural properties then raise the relevance of symmetry context to understand the physical behavior of electrons. In section exposes the symmetry importance in this work and their fundamental role in emergence of the IOA. Then focus on symmetry reduction (symmetry breaking) which is the causes of appearance of interesting physical properties, in our case optical properties.

The perturbative model purposed to understand of the IOA is simple and useful, as this depend on grade of asymmetry in the CQWs system, the more asymmetry increases the RAS signal. Also, this model is support by the numerical calculations with a good approximation. When it had obtained the firsts results, we purposed to developed codes to generate numerical results and although this represented a new area to explore, we decided to dedicated enough time to get a new tool to support our experimental work. Already inside in numerical solutions' area, we realized of complexity of generate reliable results, overall to existence of numerous proposals to get it, then we decided implemented the simplicity context. Our numerical results are simple but reliable in accordance with the numerical results, by this reason we create a [GitHub](#) repository [179] with aim to developed new codes and numerical models which in future work can will be implemented. In the experimental part, the results are the proof of the arduously work that it was inverted, along of the project it was realized experiments to study and understand the physics which involves the CQWs systems, although this has been study for several years ago, our contribution it's novel.

The quantum confinement is the key of these structures, if add the symmetry breaking by coupled wells width asymmetry exhibits wonderful physics, from spin dynamics to excitonic effects. With respect to spin dynamics into CQWs structures, it was realized experiments of circularly polarized PL over the samples shown in this work, reveals that the spin relaxation time τ_s . It has been demonstrates that the degree of circular polarization is directly related to the asymmetry of the CQWs [90].

The excitonic properties shown in the Section 3.2.2.1 are really interesting, the PR experiments performed as a laser power function reveal a non-common transition in these experiments, this transition associated with a trion, commonly occurs in structures under external disturbance as an electric field applied, in our case, we not only detect the trion transition, but also it modulated with a light source. This means this can be applied as a laser transistor. These PR results are very relevant, in fact as a future work we planned to publish them.

Finally, in the RAS experiments it's clearly the wonderful physics which exhibits ACQWs structures, the results obtained are the principle of an experiments' series which we're thinking to carried out. Without intention to being repetitive since the results proofs our

hard work, we purpose to take further the RAS technique to explore with more detail the ACQWs structures. The first upgrade of RAS experiments, it's do it spin sensitive, this means, spin resolve RAS experiments.

The principal idea is enhancing the RAS setup to measure spin response. Our proposal is to carry out the experiments just changing the modulated PEM polarization. In the RAS setup (as can see in Figure 3.19) the monochromatic beam is first polarized to then modulated linear polarization by the PEM between mutual perpendicular polarization states, to finally spot on sample along [110] and [1 $\bar{1}$ 0] directions. Then, in the spin resolve RAS experiments, it purpose to modify PEM polarization modulation, to this it's possible to choice that the modulation being right circular and left circular polarization, in the PEM device this is $\lambda/4$ and $3\lambda/2$ which in contrast with the $\lambda/2$ of the traditional RAS experiments.

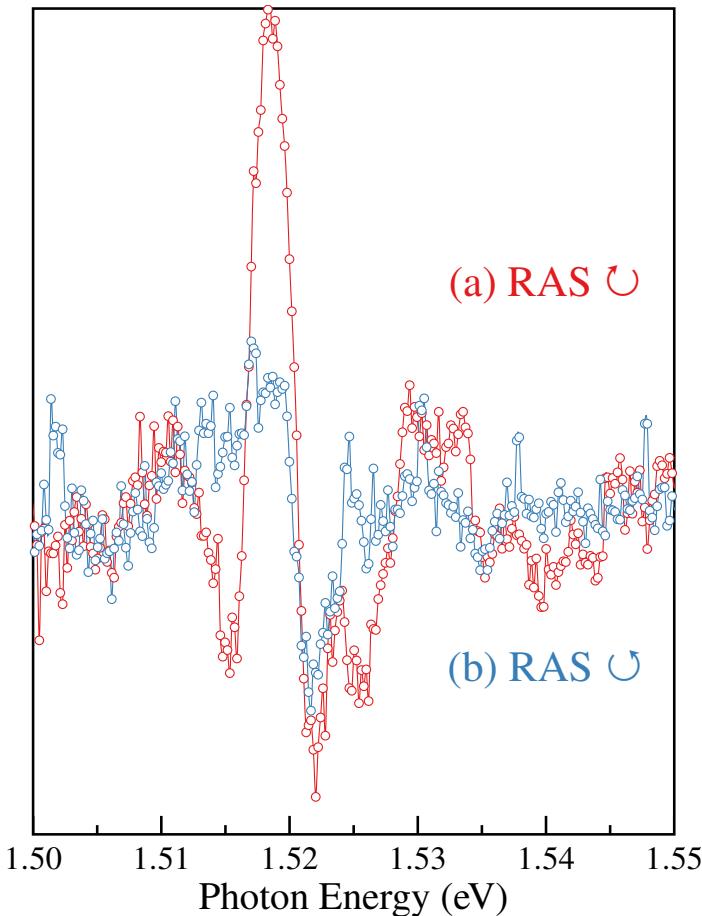


Figure 4.1: Spin resolve RAS experiments worked on ACQWs-2 sample, this experiments carried out in sequential way, firstly measured with one polarization state and then the second state. The sample it was placed along preferential direction ([1 $\bar{1}$ 0]), maybe this be a reason can observer a structural signal of RAS, this means, the line shape of anisotropy due to the asymmetry structure. Although, it's notable the difference between mutual polarization states.

The first results obtained from worked on ACQWs-2 sample shows in Figure 4.1, these experiments were it performed with the sample position in a preferential direction [1 $\bar{1}$ 0] and as a sequential way, this means, firstly with a polarization state then the other state. The results shown in Figure 4.1 are the average of sequential experiments, each of one with their respective polarization. The signals results exhibit an interest difference, although,

maybe the line-shape has a remainder of structural anisotropy due to the asymmetry in the structure (C_{2v}). If taken into account the contribution of structural anisotropy, it appears to be polarization is longer. Even if, these results are really first approximation, the technique has powerful to explore spin properties. The second RAS upgrade to spin resolve, it's a proposal, instead of polarization states with the PEM will be used a laser beam circular (left and right) focused onto the sample, while the RAS signal it's measured traditionally. We expected that these upgrades the RAS experiments, turning a tool for spin study.

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