Optical anisotropies of asymmetric double GaAs (001) Quantum Wells

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In the present work, we were able to identify and characterize a new source of in-plane optical anisotropies (IOAs) occurring in asymmetric DQWs; 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 (DQWs) structures coupled by a thin ($< 2$ nm) tunneling barrier. Two groups of DQWs systems were studied: one where both QWs have the same thickness (symmetric DQW) and another one where they have different thicknesses (asymmetric DQW). RAS measures the 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 DQW is 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 asymmetric DQW 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 loses 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 DQWs.

I. INTRODUCTION

Nanostructures based on double quantum wells (DQWs) constitute an excellent platform to study the interactions between confined energy levels; for instance, intra-QW transitions between electrons and holes belonging to the same QW (direct excitons and trions), and inter-QW transitions occurring for electrons and holes resident in different QW (indirect excitons and indirect trions) [1]. The strength of the direct and indirect transitions can be modulated by the application of external perturbations, such as external electric fields. The electric field modifies the tunneling probability leading to the appearance of spatially indirect inter-QW excitons [1, 3] and intra-QW charged excitons (trions) [1]. Such complex band structures offer interesting optical phenomena which exhibit polarization dependence. In-plane optical anisotropies (IOAs) of AlGaAs/GaAs-based heterostructures have been studied extensively in the past several years by means of reflectance anisotropy spectroscopy (RAS). In the case of single QWs, several sources of IOAs have been investigated: i) absence of 4-fold rotational symmetry axis at the AlGaAs/GaAs interface [4, 5], ii) asymmetric quantum wells [6], iii) nonuniform segregation of Ga atoms at AlGaAs/GaAs interfaces [5, 7], iv) application of an external stress [8, 9], and v) asymmetric barriers [10]. In general, the observed IOAs strength depends on the QW thickness. For example, for strain-induced IOAS, the RAS response increases with the well thickness [8, 9]; whereas it decreases with well thickness for the anisotropies induced exclusively by the interfaces [5, 8, 10].

In DQW-based systems, the relative thickness of the QWs plays an important role in the anisotropy strength; namely, if both QWs have the same thickness (symmetric DQW), the IOA arises from the contributions of the AlGaAs/GaAs(QW) and GaAs(QW)/AlGaAs interfaces which, in general, are not equivalent. If the QWs thicknesses are different (asymmetric DQW), the system loses its inversion symmetry as a whole along a plane perpendicular to the growth direction an passing through the center of the tunneling barrier; therefore, the interfaces of the DQW system are not distributed symmetrically at both sides of the tunneling barrier, leading to an increase of the IOA signal.

In this paper we report on RAS experiments on DQWs coupled by a thin barrier (1.98 nm) and different relative thicknesses. We find that by increasing the
TABLE I. Structure of samples used in this work.

| Sample number | Well width $d_1$ (nm) | Well width $d_2$ (nm) | Si$_{0.15}$Ga$_{0.85}$As |
|---------------|----------------------|----------------------|------------------------|
| S1            | 11.87                | 11.87                | Si doped, $n = 6 \times 10^{18}$ cm$^{-3}$ |
| A1            | 13.85                | 11.87                | Si doped, $n = 6 \times 10^{18}$ cm$^{-3}$ |
| A2            | 23.74                | 11.87                | Si doped, $n = 6 \times 10^{18}$ cm$^{-3}$ |
| A3            | 23.74                | 11.87                | Be doped, $p = 5 \times 10^{18}$ cm$^{-3}$ |

thickness of one of the QWs relative to the other, we induce a breakdown of inversion symmetry of the DQW structure which increases the IOA.

The rest of the paper is organized as follows. In Sec. II a description of samples and the experimental setup is given. In Sec. III a description of the numerical model used to calculate the energy of the excitonic transitions of the DQWs and a detailed description of the model used to explain the RAS spectra are discussed. In Sec. IV we describe the RAS and photoluminescence (PL) on symmetric and asymmetric DQWs as well as their interpretation on the basis of the breakdown of inversion symmetry. Finally, conclusions are given in Sec. V.

II. SAMPLES AN EXPERIMENTAL SETUP

The samples were grown by molecular beam epitaxy (MBE) on a GaAs (001) semi-insulating substrates covered with a 200 nm-thick smooth GaAs buffer layer (see Fig. 1 and Table I). On top of this layer, a 600 nm-thick, n-type (samples S1, A1, A2) or p-type (sample A3) Si$_{0.15}$Ga$_{0.85}$As layer was grown. The DQW consists of two undoped GaAs QWs of thicknesses $d_1$ and $d_2$ separated by a 1.98 nm thick Si$_{0.15}$Ga$_{0.85}$As barrier. Sample S1 is a symmetric DQW (i.e., with $d_1 = d_2$) while samples A1, A2 and A3 are asymmetric ($d_1 \neq d_2$). The whole structures were covered with a 10 nm-thick GaAs capping layer.

RAS and reflectance (R) experiments were carried out using for excitation the monochromatic probe light obtained by filtering a 70 W tungsten lamp with a monochromator. The monochromatic beam then passes through a polarizer prism and a photo-elastic modulator in tandem and is focused onto the sample with a spot size of 5.0 mm of diameter. The sample is chilled at cryogenic temperatures (30 K) by means of a Helium closed cycle cryocooler. The light reflected by the sample is collected and detected by a multialkali photomultiplier tube. A detailed description of the RAS technique can be found elsewhere [11].

III. MODEL

A. Numerical calculations

The one-dimensional Schrödinger equation in the effective mass approach was solved using the finite difference method [12, 13] to obtain both the transition energies and wave functions of the DQWs. We neglect spin-orbit interaction, which leads to a 4x4 Luttinger’s Hamiltonian [14] with decoupled heavy- and light- hole states. Taking into account the band-edge variation $V(z)$ as well as the dependence of the effective mass $m(z)$ on the composition of the layers [13, 15, 17], our system can be described...
by:

$$-\frac{\hbar^2}{2m_jz} \frac{d^2}{dz^2} + V(z) \psi_{nj}(z) = E_{nj} \psi_{nj}(z),$$

$$j = e, hh, lh.$$

The Varshni equation was employed for the determination of gap energies of Al$_x$Ga$_{1-x}$As alloys [13]. The conduction and valence band offsets were chosen as 65% and 35%, respectively. Both electron and hole effective masses for GaAs and AlAs can be found in Refs. [19–21], whereas for Al$_x$Ga$_{1-x}$As the Vegard’s law was used [22].

Figure (2) shows the energies and wave functions of the S1 (Fig. 2(a) and A1 (Fig. 2(b)) samples calculated by the finite difference model without band nonparabolicity. The transition energies were calculated by taking into consideration the exciton binding energy as a function of the well width [16, 23]. We show that wave functions in asymmetric DQW are asymmetrically distributed around the tunneling barrier for both electrons and holes. The barrier width plays an important role in tunneling. For symmetric DQW, as noted above, the electron and heavy- light- holes states are quite similarly localized in the two wells and there is a splitting of the degenerate single states into doublets. For asymmetric DQW the energies of doublets tend to the singlet state energy of a single QW [24].

B. Anisotropy Model

GaAs is a cubic crystal of the symmetry group $T_d$. For a single QW grown on the (001) surface, the $z_\parallel (001)$ direction becomes inequivalent to both $x_\parallel (100)$ and $y_\parallel (010)$ directions, hence lowering symmetry from $T_d$ to $D_{2d}$. This group comprises eight operations of symmetry including four that change the direction of the $z$-axis. In this symmetry group, the $x$ and $y$ axes remain equivalent and the optical properties are isotropic in the $x - y$ plane. An IOA is expected when the $z_\parallel (001)$ axis loses its 4-fold rotation symmetry, lowering the symmetry further from $D_{2d}$ to $C_{2v}$. Different perturbations have been considered to increase the IOA; among them we can count on: uniaxial external stress applied along [110] direction, ultra-thin layers inserted in one QWs barrier, and external electric fields applied along the $z_\parallel (001)$ direction. Another mechanism that induces an IOA in a single QW is the non-equivalence of the AlGaAs/GaAs interfaces. As a matter of fact, in a single QW the anisotropies originated at the AlGaAs/GaAs(QW) and at the GaAs(QW)/AlGaAs interfaces are reversed in sign, thus cancelling the IOA ($D_{2d}$ symmetry). However, it is well-known, that, experimentally, they are not exactly equivalent from the structural point of view [9] and a residual IOA is normally observed [4, 5] ($C_{2v}$ symmetry). RAS spectra of single QWs have been reported extensively in the literature [5, 6, 8–10].

The thickness of the QW plays a fundamental role in the amplitude of the RAS signal. For the anisotropies originated 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 [5]. Contrary to this behavior, for the IOA induced by strain, the RAS signal increases with the thickness of the QW [8]. In any case, the RAS signal is associated to the mixing between the heavy- and light-hole valence subbands of the QW [23]. In a perturbative approach the IOA strength is proportional to:

$$\Delta E_n = \frac{\langle \psi_{en} | H | \psi_{hhn} \rangle \langle \psi_{hhn} | H | \psi_{en} \rangle}{\langle \psi_{en} | \psi_{hhn} \rangle}$$

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

![Figure 2](image-url)
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 coupled DQW system, $\mathcal{H}$ is a measure of the asymmetry of the two QWs. In a DQW, 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 QWs, as can be seen in Fig. 2(a). 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 are non-equivalent, and a residual IOA is induced by the symmetry reduction from $D_{2d}$ to $C_{2v}$ of the whole DQW 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 (asymmetric DQW), 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 Fig. 2(b). Besides of the anisotropic source induced by the GaAs/AlGaAs interfaces as in the symmetric case, for the asymmetric DQW system another mechanism of IOA should be considered. Suppose that the AlGaAs/GaAs interfaces are equivalent. As we mentioned, in this case a symmetric DQW 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 Fig. 2(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 asymmetric DQW structure is reduced to the four operations of the symmetry group $C_{2v}$ [26, 27]. In this case the IOA strength can be modulated by changing the relative thicknesses of the QWs in the DQW system.

This anisotropy is added to the residual IOA produced by the non-equivalence of the interfaces.

### IV. EXPERIMENTAL RESULTS

Figure 3 shows the RAS spectra recorded at 30K for the samples S1, A1 and A2 (described in Table I). The inset plot show the corresponding PL spectra measured at 12K, where the excitonic transitions can be identified by two peaks, the larger one associated with e1-hh1 transition and a much smaller one associated to the e1-lh1. In the case of RAS, the peaks associated with these transitions are seen clearer. The e1-hh1 and e1-lh1 transitions determined in the PL spectra are indicated by arrows. These values are in excellent agreement with those determined by the numerical calculations (see Table I). Note that the features associated to each transition have opposite concavities and red-shift when one of the QWs of the system increases its thickness. The evolution of the optical structure is evident: if the DQW structure become asymmetric the corresponding RAS spectra increases its strength.

Due to the fact that the wave function probability density of sample S1 is distributed symmetrically along the QWs 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 $0.1 \times 10^{-3}$ has been reported for a 8 nm single QW [3], which has the same order of magnitude as in spectra of Fig. 3(c). This IOA is attributed to the inequivalent AlGaAs/GaAs interfaces along the DQW structure.

As pointed out before, the strength of the IOA signal is produced by an intermixing of the heavy- and light hole states in the valence band which is proportional to $\langle \psi_{\text{hhn}} | H | \psi_{\text{hhn}} \rangle$ according to Eq. (2). For the lowest heavy and light holes 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 A2 (A3), A1 and S1, respectively. The mixing $\langle \psi_{\text{hhn}} | H | \psi_{\text{hhn}} \rangle$ can be estimated by considering that the transitions are direct ($n$ is the

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### Table II. Comparative of experimental ($E$) and numerical calculations ($N$) of transition energies (in eV). $\delta E_e$, $\delta E_{hh}$ and $\delta E_{lh}$ corresponds to the difference between electrons, heavy- light holes states, respectively. $\Delta E_n$ is the numerical calculation of energy splitting for transitions 1 and 2 ($n = 1, 2$).

| Sample | e1-hh1 | e1-lh1 | e2-hh2 | e2-lh2 | e3-hh3 | $\delta E_{eV}$ | $\delta E_{hhV}$ | $\delta E_{lhV}$ | $\Delta E_n$ ($meV$) |
|--------|--------|--------|--------|--------|--------|--------------|--------------|--------------|----------------|
| S1     | (E)1.5328 (N)1.5297 | (E)1.5380 (N)1.5341 | (N)1.5394 (N)1.5499 | (N)1.5948 | 8.9 | 0.9 | 6.9 | $\Delta E_1 = 4.4$ |
| A1     | (E)1.5265 (N)1.5273 | (E)1.5296 (N)1.5314 | (N)1.5368 (N)1.5460 | (N)1.5837 | 8.4 | 1.3 | 6.4 | $\Delta E_1 = 4.1$ |
| A2     | (E)1.5181 (N)1.5190 | (E)1.5206 (N)1.5210 | (N)1.5330 (N)1.5394 | (N)1.5460 | 11.8 | 3.4 | 7.5 | $\Delta E_1 = 2.0$ |
FIG. 3. RAS spectra for the (a)-(b) asymmetric and (c) symmetric DQWs. 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 to the transition e1-hh1 and a much smaller one associated to the e1-lh1 (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 to e1-hh1 and e1-lh1 increase their strength when the DWQs becomes more asymmetric. The RAS spectra were measured at 30K.

FIG. 4. Reflection anisotropy (RAS, open and closed circles) and differential reflection (DR, solid line) spectra for asymmetric DQWs A2 and A3, grown on an AlGaAs n-type and p-type layer respectively. Note that while for the heavy hole transitions (e1-hh1 and e2-hh2) in the RAS and DR spectra have the same concavity, for light holes transitions (e1-lh1 and e2-lh2) 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.
The arrows at the bottom of Fig. 4 indicate the energy of the states en-lhn 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 1, the energy splitting between transitions en-lhn and en-lhn are $\Delta E_1 = 2.0 \text{ meV}$ and $\Delta E_2 = 6.4 \text{ meV}$. In accordance with Eq. 2, the IOA amplitude is proportional to $1/\Delta E_m$. Considering the same valued for the overlapping and the mixing $\langle \psi_{lhn} | H | \psi_{lhn} \rangle$ 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 Fig. 4 supporting our interpretation.

Finally, we discuss the possible contribution to the RAS amplitude by an built-in electric field across the DQW. To study this contribution, we have compared the RAS spectra of asymmetric samples A2 and A3. The difference between them is the doping of the AlGaAs layer (see Fig. 4). While for A2 it is n-type, for A3 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 A2 and A3. Thus, the linear contribution of the electric field to the RAS should be reversed in sign for such samples. Figure 4 shows the comparison between the RAS spectra of asymmetric samples A2 and A3 obtained from the difference in reflectivity between the [110] and [110] crystallographic directions. As can be seen, 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. We conclude, thus, that the dominant contribution to RAS spectra is the asymmetry of the DQW system.

V. CONCLUSION

We presented in this work a detailed study of the evolution with QW thickness of the RAS spectra of a coupled DQW system. The RAS signal increases its strength with the degree of asymmetry of the QWs that constitutes the DQW system. This behavior is attributed to the reduction of the symmetry of the electronic states in the DQWs with QWs of different thicknesses. The nature of the transitions was identified by using PL spectroscopy and numerical calculations. We believe that the result presented in this work are important and will be useful in understanding of the evolution of the optical transition induced by the breakdown of translation symmetry in asymmetric DQWs.

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