Line-shape analysis of the reflectivity spectra of GaAs/(Ga,Al)As single quantum wells grown on (001)- and (311)-oriented substrates

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The reflectivity line shape of single quantum wells is calculated as a function of the thickness of the sample structure near the excitonic resonances. By direct comparison between the calculation and the experimental reflectivity we can deduce the oscillator strength of 1*s* and 2*s* excitons in the case of $GaAs/Ga_{0.7}Al_{0.3}As$ single quantum wells grown on (001)-oriented substrates. $GaAs/Ga_{0.81}Al_{0.19}As$ single quantum wells grown on (311)-oriented GaAs substrates with thickness ranging from 5 monolayers up to 87 monolayers have been also studied. The light-hole exciton is found to be more sensitive to the growth direction than the heavy-hole one.

I. INTRODUCTION

Since the advent of quantum wells, a lot of theoretical and experimental investigations have been devoted to the determination of the excitonic Rydberg energy as a function of the quantum-well design (i.e., confining layer thickness and barrier heights) in case of (001)-grown quantum wells.[1] Similar studies have not been so extensively made when growth occurs in a direction lying far away from a high-symmetry direction of the zinc-blende lattice. To the best of our knowledge, only the (111), (110), and (310) orientations have been investigated.[2–4] The contribution of (311) orientation to this field is addressed in the present paper. Using a high-quality (001)-grown $GaAs/Ga_{1-x}Al_xAs$ single quantum well, we first check the theoretical method employed to calculate the change in reflectance near the excitonic resonances in single quantum wells. In particular, from the fit of the experiment, we obtain the oscillator strengths of 1*s* and 2*s* excitons and compare this result with the values theoretically predicted.[5,6] Both photoluminescence (PL), photoluminescence excitation (PLE) spectroscopy, and reflectivity line-shape analysis (RLSA) patterns have been chosen as experimental methods. The joint density of states between conduction and valence subbands can be sensitive to the growth orientation due to the anisotropy of the dispersion energy in the valence band of bulk materials; this may have an importance in device engineering.[2] Changing the growth orientation first requires us to perform a material-science exercise in order to grow good enough quality samples; secondly, it leads to complications for calculating the Rydberg energies when strong anisotropy of the valence-band mass has to be included in the theory. In this work, the Rydberg energy has been deduced by subtracting the experimental transition energy from the calculated ("Rydberg-less") one. The net result obtained for quantum-well widths ranging from 24.4 down to ~ 1.5 nm is a stronger influence of the growth orientation for light-hole excitons (LHE's) than for heavy-hole excitons (HHE's).

II. EXPERIMENTAL DATA

A. Experimental setup

PL and PLE studies are performed at 2 K, using a front-surface layer excitation by a tunable dye laser with Styryl-11 and Styryl-12 dyes pumped with an argon-ionized laer. PL and PLE are analyzed by a 1-m focal-length double monochromator and detected by photon counting using a photomultiplier tube with a cooled GaAs photocathode. Reflectivity measurements were made using another experimental setup where the focused image of a 100W tungsten-wire lamp was reflected onto the input split of a 1.5-m focal-length monochromator, and dispersed and detected with a GaAs photomultiplier followed by a lock-in amplifier.

B. Sample characteristics

The (311)-grown sample was grown by molecular-beam epitaxy (MBE) at the Optoelectronic Joint Research Laboratory (Kawasaki, Japan). It consists of five single quantum wells (SQW's) isolated from one another by 45-nm-wide (Ga,Al)As barrier layers and a 5-nm-wide GaAs cap layer. Details concerning the growth can be found elsewhere.[7] The well widths were estimated from the growth time of each well and were found to be ranging from 5 up to 87 monolayers. The (001)-grown sample consists of

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one 79-Å-wide GaAs layer sandwiched between one 110-nm-wide $Ga_{0.68}Al_{0.32}As$ front-barrier layer and a 50-nm-thick back-barrier layer, without a cap layer. This sample was grown by MBE at Laboratoire de Physique du Solide et Energie Solair (Valbonne, France) and the well width was measured by the reflection high-energy electron diffraction (RHEED) oscillation method.

III. THEORETICAL METHODS

Let us consider the reflection of an electromagnetic wave at the interface between two media with different refractive indexes n_α and n_β. Under normal-incidence conditions, the reflectance R is given by

$$R=|(n_\alpha-n_\beta)|^2/|(n_\alpha+n_\beta)|^2 .$$

To adapt this result to the case of GaAs/(Ga,Al)As single quantum wells, we extend the ideas developed in Ref. 8. Reflectance results were gathered of interference between beams reflected at the air-sample interface, at the front-barrier–GaAs-layer interface, at the GaAs-layer–back-barrier interface, and at the back-barrier–GaAs interface. Let r_0, r_1, r_2, and r_3 be the respective amplitudes of these beams. The reflectance R is obtained by the square of the modulus of the complex quantity $(r_0+r_1+r_2+r_3)$ which can be calculated as a function of the refractive index of the different layers, taking into account the phase angles δ_1, δ_2, and δ_3 of the optical path in the front-barrier, quantum-well, and back-barrier layers. We write

$$\delta_\alpha=2\pi n_\alpha d_\alpha/\lambda ,$$

where d_α is the thickness of the layer for the α compound. δ_α is a real quantity for a transparent medium; it becomes a complex quantity when an absorption occurs. Then the absorption is given by the imaginary part of δ_α. The amplitudes r_1, r_2, and r_3 are obtained using a model for the dielectric constant which we detail below.

For the (311)-grown sample, because the thickness of the front barrier varies from a given SQW to another, drastic changes of the reflectivity line shape are expected, due to modification of the interference systems mainly due to the variations of δ_1.[8] In this calculation, given a quantum well, an effective barrier thickness is taken as the sum of the thicknesses of the GaAs cap layer, the alloy barrier thicknesses, and thicknesses of thinner GaAs wells. For simplicity, in what follows, the dielectric constant in the front barrier is then calculated as the weighted means of the GaAs and $Ga_{1-x}Al_xAs$ barrier dielectric constants ϵ_0 and ϵ_1. The relationship between the thickness of the GaAs layers and the band gap of the corresponding quantum well support this approximation: at the energy of the quantum well under consideration, thinner quantum wells are transparent. In the GaAs layer the dielectric constant is taken as a sum of standard oscillators centered at the energy of the excitonic resonances.[8] We write

$$\epsilon=\epsilon_0+\frac{A_{HH}}{\omega^2-\omega_{HH}^2-i\omega\Gamma_{HH}}+\frac{A_{LH}}{\omega^2-\omega_{LH}^2-i\omega\Gamma_{LH}} . \quad (1)$$

A_{HH} and A_{LH} are related to the oscillator strengths for heavy- and light-hole-exciton transitions centered at energy ω_{HH} and ω_{LH}, respectively; the Γ_{ij} quantities are the corresponding broadening parameters. The oscillator strengths are expected to increase when the quantum-well thickness decreases, while they are reduced in the small thickness range when the two-dimensional (2D) character of the heterostructure vanishes.[4,5] Therefore, a close correlation between the light-hole–heavy-hole splitting and the magnitude of both oscillator strengths is also expected.[9–11] Selection rules for the band-to-band transition between conduction- and valence-band Bloch waves suggest a smaller oscillator strength for a light-hole exciton than for heavy-hole one (in a ratio of ~ 0.33).[12] Hybridization effects between valence subbands and couplings between, for instance, $3d$- and $1s$-exciton envelope functions may also have a noticeable influence.[4,5,13] Taking into account the contribution of the excitonic excited states requires us to extend the summation in (1) using additional oscillators.

IV. RESULTS

A. (001)-grown sample

The aluminum concentration in the barriers has been obtained from the reflectivity structure at ~ 1.95 eV; we find $x=0.32$ according to the gap-composition relation given in Ref. 14. Figure 1 displays the experimental

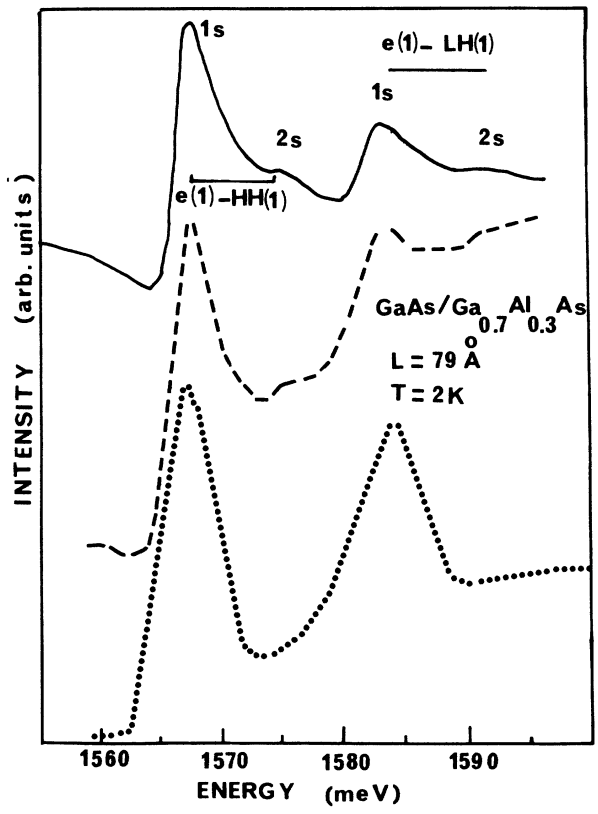

FIG. 1. Optical properties of $GaAs/Ga_{0.7}Al_{0.3}As$ single-quantum-well–grown on a (001)-oriented substrate. Dotted lines correspond to the photoluminescence-excitation-spectroscopy data, dashed lines to the experimental reflectivity, and the solid line is the result of the calculation.

reflectivity spectrum (dashed line) and the PLE one (dotted line) which was collected by setting the spectrometer at 1.566 meV. Fitting the reflectivity curve is not possible without introducing additional oscillators to account for the ~1575- and ~1591-meV structures. When this is done, we can obtain the theoretical spectrum (solid line) using $A_{HH}=12000$ meV^2, $A_{LH}=5000$ meV^2, $\Gamma_{HH}=3$ meV, $\Gamma_{LH}=4$ meV, $\omega_{HH}=1566.44$ meV, and $\omega_{LH}=1581.78$ meV for the two main transitions. The values obtained for the weaker structures are $A'_{HH}=A'_{LH}=600$ meV^2, $\Gamma'_{HH}=3$ meV, $\Gamma'_{LH}=4$ meV, $\omega'_{HH}=1574.66$ meV, and $\omega'_{LH}=1690.82$ meV. We now discuss these quantities. The ratio A_{HH}/A_{LH} equals 2.4. It closely fits the ratio of oscillator strengths calculated for 1s heavy-hole and 1s light-hole excitons in Refs. 4 and 5. The 8.22-meV splitting between the two lower-energy structures agrees with an identification in terms of 1s and 2s heavy-hole excitons; for light-hole excitons, we find 9.04 meV. This is consistent with the results obtained for other aluminum concentrations, which are summarized in Ref. 1. The most important result concerns the oscillator strengths; we find $A_{HH}/A'_{HH}=20$ and $A_{LH}/A'_{LH}\sim 8.3$. Our fitting procedure gives $A'_{HH}=A'_{LH}$. The relationship $A_{\alpha\beta}=4\pi\Phi_{\alpha\beta}\omega^2_{\alpha\beta}$ between oscillator strength $\Phi_{\alpha\beta}$ and our fitting parameter $A_{\alpha\beta}$ suggests an oscillator strength for 2s light-hole excitons smaller than the one of 2s heavy-hole excitons.

B. (311)-grown sample

Figure 2 displays the general photoluminescence spectra of the five SQW's grown along the (311) direction. The corresponding photoluminescence-excitation data collected setting in each case the spectrometer at the energy of the photoluminescence maximum have been reported in Figs. 3–6 (dotted lines). In the PLE spectra of Fig. 6, the onset of the $Ga_{1-x}Al_xAs$ band gap is responsible for the ~1.8-eV transition. The aluminum concentration was estimated to be 0.19.[11] The experimental reflectivity data are reported in Figs. 3–6 (dashed lines). Neither PLE nor R spectra enabled us to observe 2s-related excitonic transitions. This can be related to the characteristics of the interfaces and the low aluminum concentration in the barrier.

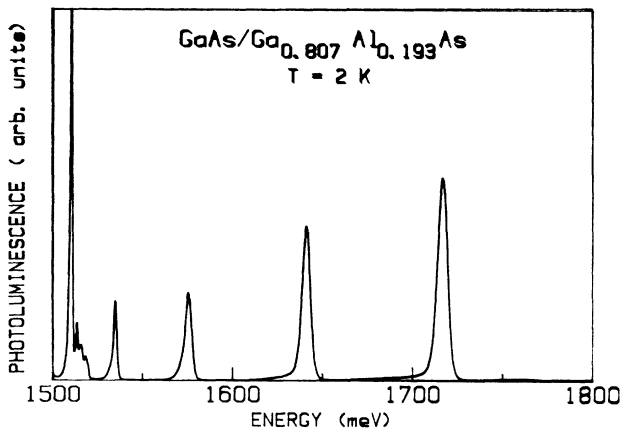

FIG. 2. 2-K photoluminescence spectrum of the five single quantum wells grown on a (311)-oriented substrate.

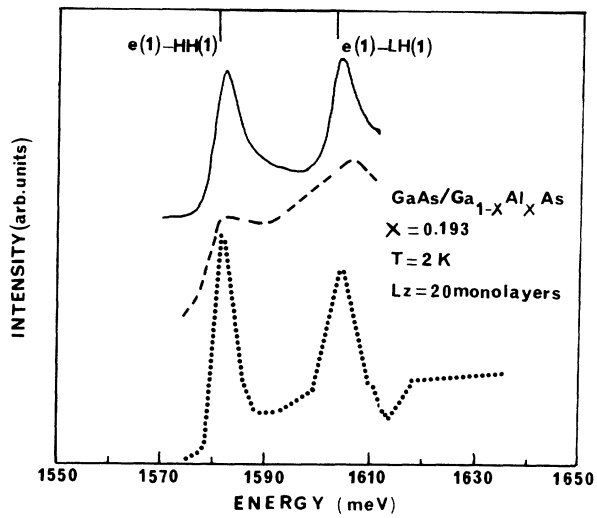

FIG. 3. Photoluminescence-excitation-spectroscopy data (dotted lines) of the two wider (311)-grown SQW's. The spectrometer was set to the maxima of the photoluminescence peaks. The experimental reflectivity is given by dashed lines; the solid lines correspond to the calculation. Note the change of the reflectivity line shape with the depth of the confining layer in the sample.

In Figs. 3–6 the comparisons between calculated (solid lines) and measured reflectivity patterns (dashed lines) are summarized. The influence of various front-barrier thicknesses modifies the reflectance shape through changes of the phase angles δ_1, δ_2, and δ_3 of the optical path in the barriers and the confining layer. As a consequence, the transition energies do not always correspond to the minima of the reflectance structures. This is illustrated on the top of the frame of Figs. 3–6, where vertical lines indicate the energies of the oscillators obtained after the fitting of the experimental data. The theoretical

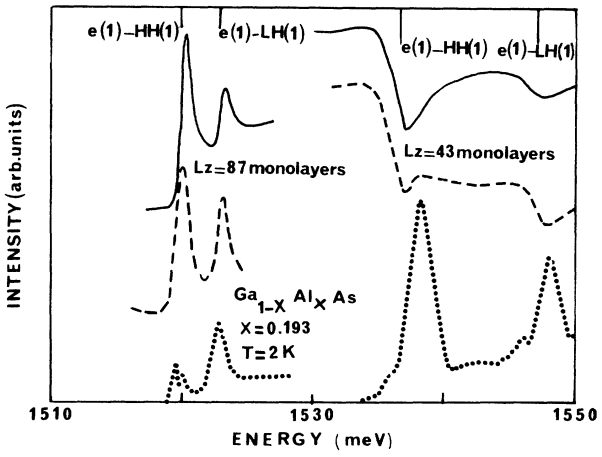

FIG. 4. The same as Fig. 3 but in case of a narrower SQW. The reflectivity structures exhibit an "absorptionlike" shape.

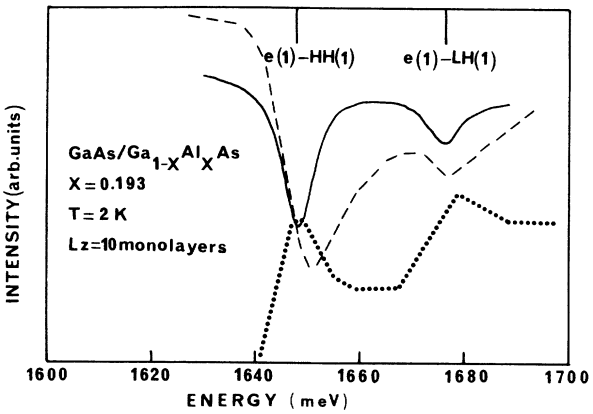

FIG. 5. A summary of the experimental data collected for this fourth SQW.

calculation requires two adjustable parameters for each excitonic transition: A_{ij} and Γ_{ij}. Table I summarizes the results of our fitting procedure. The variation of the damping parameter as a function of the quantum-well width roughly follows the change of the splitting between confined valence levels and the width of the PLE peaks. Such a trend has also been obtained for the oscillator-strength parameters A_{ij}, but the ratio between deduced values of A_{HH} and A_{LH} was not found to be constant for our series of SQW widths. This last finding is not actually understood, but has to be carefully considered because we cannot presently say if it has a physical origin or results from our overly simplified treatment of the reflectivity in SQW's: comparing with the theoretical calculation of reflectivity in semi-infinite semiconductors, it is obvious that the analog of the so-called additional boundary conditions has been omitted in our calculation.[15–17] The damping parameters obtained for the (001)-grown sample are ~3–4 meV, a value which may be inserted for comparison in Table I between rows corresponding to well widths of 20 and 43 monolayers. The (001)-growth value is comparable to the value we could expect by extrapolation of the (311) data of Table I; this enables us to conclude that the quality of the interfaces

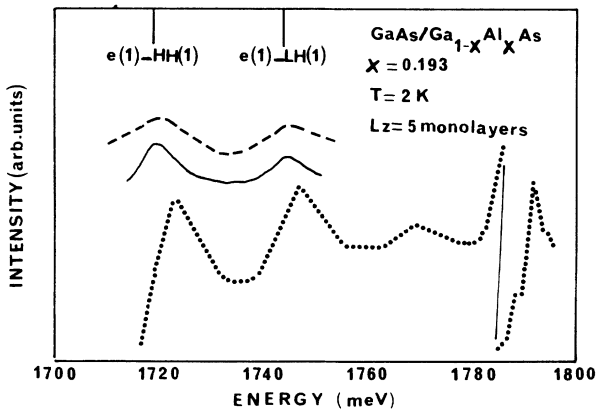

FIG. 6. The same as Figs. 3–5. The onset of the (Ga,Al)As band gap at 1793 meV has been given on the PLE data.

TABLE I. Parameters obtained from the fitting of the reflectivity spectra for the (311)-grown sample.

Well width (monolayers)	A_{HH} (meV^2)	A_{HH} (meV^2)	Γ (meV)
5	7500	4500	9.0
10	20000	8000	7.5
20	10000	7000	5.0
43	2600	1000	2.5
87	2500	900	0.8

obtained in case of (311) growth is reasonable.

To estimate the (311)-related Rydberg energies, we have calculated the confinement energy using the envelope-function approach. The hole quantization masses have been taken following Eppenga *et al.*[18] Subtracting the experimental transition energy from the calculated one, we estimate the Rydberg energy. The dependence of light- and heavy-hole Rydbergs as a function of the layer thickness has been plotted in Fig. 7 (circles correspond to RLSA data, and squares correspond to PLE data). Dashed lines are guides to the eye. The scattering between RLSA and PLE increases when the well width decreases, and this is when the influence of interface quality becomes important. The thickness of the confining potential is found to have a stronger influence on the light-hole exciton energy than on the heavy-hole one. Comparing our values of the effective Rydberg with the values previously obtained for this range of aluminum concentration in the barriers and when growth occurs in the (001) direction,[19] we find that the light-hole-exciton Rydberg is more sensitive to the substrate orientation than the heavy-hole one. Such an effect agrees with previous observations for (110)-, (111)-, and (310)-grown SQW's.[6,20]

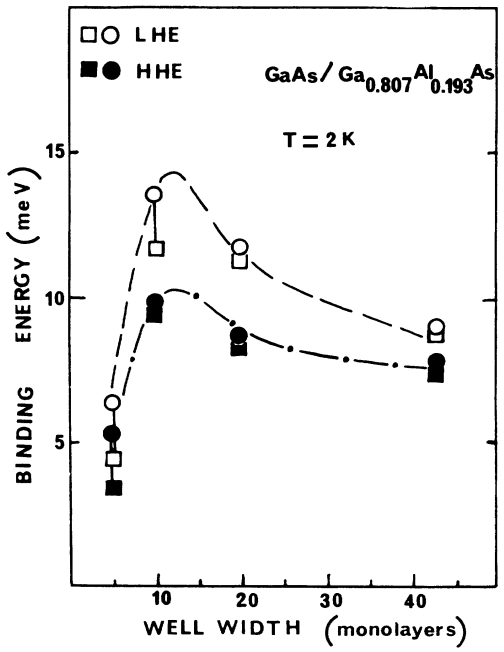

FIG. 7. Variations of the effective Rydbergs obtained for the (311)-grown sample. Dashed and dashed-dotted lines are guides to the eye.

V. CONCLUSION

We have reported the direct measurement of oscillator strengths for 1*s* and 2*s* heavy-hole and light-hole excitons in case of a (001)-grown GaAs/$Ga_{0.7}Al_{0.3}As$ single quantum well. Our finding is in agreement with previous theoretical calculations. PLE and RLSA have been performed at 2 K on a series of (311)-oriented GaAs/(Ga,Al)As single quantum wells. A good reflectivity investigation could be made for layer thicknesses of 5 monolayers. Comparing the line shape of the reflectivity together with the one we calculated using a simple "classical" model, we were able to propose an experimental trend for the LHE and HHE binding energy in narrow (311)-oriented GaAs/(Ga,Al)As single quantum wells. This treatment of the reflectivity gives Rydberg values which agree with the photoluminescence-excitation-spectroscopy data within what we believe to be the experimental uncertainty.

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