Uniaxial-stress investigation of asymmetrical GaAs-(Ga,Al)As double quantum wells

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We present low-temperature-reflectance experiments, performed on GaAs-$Ga_{1-x}Al_xAs$ asymmetrical double quantum wells, under in-plane uniaxial stress. The results, when compared to what is obtained from single quantum wells, exhibit some peculiar behaviors, which are typical effects of the asymmetry of the structures. A careful examination of these results is proposed in light of theoretical predictions about subband-to-subband transition energies and oscillator strengths. Our conclusion is that asymmetry makes it possible to obtain novel couplings between excitonic states, which cannot be accounted for without an accurate theoretical treatment of the Coulombic interaction, including intersubband mixings of valence wave functions.

I. INTRODUCTION

Resonant tunneling of carriers in multiple quantum wells is currently under very active investigation, mainly due to the fact that, owing to recent progress in growth techniques, researchers now have the opportunity to tailor the conditions under which the tunneling process can be initiated. In particular, the possibility of obtaining sharp and quick switches of the optical properties of materials, around a critical value of the applied electric field, may lead to future device applications in optoelectronics.[1] The most elementary system in which optical studies of nonresonant or resonant tunneling can be performed consists of a pair of coupled quantum wells. Quite a large number of both theoretical and experimental works on this subject can be found in the literature.[2–27] These works mainly concern studies of the canonical system GaAs-(Ga,Al)As. However, progress in the control of interface qualities for heterostructures made from combinations of more exotic compounds—in particular, those lattice-mismatched to each other—has led workers to anticipate the realization of a new generation of resonant-tunneling-based devices in the near future. Many previous works[2–14] were devoted to understanding the mechanisms of tunneling of electrons and holes, while ignoring the role played by the Coulomb interaction. This latter effect was recently the focus of a few experimental and theoretical studies.[15–27]

The purpose of the present paper is to study the evolution of the electronic structure of GaAs-(Ga,Al)As double asymmetrical quantum wells (DAQW's) subjected to external uniaxial stresses of varying intensity. The stress is applied in crossed configuration to the space-quantization field of the DAQW's. The corresponding symmetry breaking has two principal effects: (i) it changes the valence-band mixings, and (ii) it modifies the spreading of the envelope functions between the two wells, when the stress is changed.

We show that, under strain, the excitons can change their character from oblique or *interwell* (electron and hole wave functions peaking in different wells) to vertical or *intrawell* (electron and hole wave functions peaking in the same well), or vice versa. This strain-induced effect could be valuable in the engineering of strained-layer DAQW's grown on buffer layers with lattice parameters chosen so as to "tune" the strain state in the heterostructure.

This paper is organized as follows: In Sec. II, we describe the experimental data taken on a pair of GaAs-(Ga,Al)As DAQW's designed so as to give different tunneling schemes for the electrons and holes. We analyze the change of the excitonic energies obtained by reflectance spectroscopy under stress. The experimental findings are compared to previous results obtained when studying isolated QW's. Our samples show novel behaviors, typical of the asymmetrical nature of their active part (confining layers of different thicknesses). The theoretical approach used to interpret the data is also described; we compare its predictions with experiment. Finally, some conclusions concerning the physics of excitons in DAQW's are addressed.

II. EXPERIMENTS

A. Samples and experimental setup

The samples investigated are two GaAs-$Ga_{1-x}Al_xAs$ DAQW's, with a nominal aluminum mole fraction $x \simeq 0.30$ in the barriers. They were grown by molecular-beam epitaxy (MBE) on GaAs substrates, with the

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TABLE I. Physical characteristics of the samples investigated in this work. The widths are expressed in monolayers (1 ML = 2.83 Å). The aluminum composition x was nominally 0.3; the values deduced from reflection high-energy electron-diffraction measurements are shown.

Sample	L_1 (ML)	h (ML)	L_2 (ML)	x(Al)
C	35	15	13	0.33
D	33	7	12	0.29

growth axis along a [001] direction. Their characteristics correspond to those of samples C and D of Ref. 21, where details on the growth process are available. For clarity, these characteristics are reproduced in Table I. In both cases, a wide GaAs quantum well ($L_1 \sim 10$ nm) is coupled to a narrow one ($L_2 \sim 4$ nm), across a thin ($h \sim 2$ or 4 nm) $Ga_{1-x}Al_xAs$ barrier.

Reflectance spectra were taken at the pumped-liquid-helium temperature ($T = 2$ K), using the broadband light of a 100-W tungsten-filament bulb. The incident light was not focused onto the surface of the samples, so as not to induce the presence of too many photocreated carriers, which could provoke internal electric fields,[10,27] due to the asymmetry of the DAQW's. The reflected beam was focused onto the entrance slit of a Jobin-Yvon 1.5-m-focal-length grating monochromator. The spectral resolution was lowered to ~ 0.8 Å (i.e., ~ 0.17 meV, for photons of ~ 1.6 eV), which is far beneath the separation between the lines usually observed in semiconductor microstructures. The light was then dispersed and detected, in the standard synchronous mode, by a thermoelectrically cooled GaAs photocathode and an EG&G model 5207 lock-in amplifier. The experimental data were numerically treated in order to eliminate the optical response of the setup, obtained by performing the same reflectance experiment on a metallic mirror.

In order to apply a uniaxial stress of varying magnitude, the samples were cleaved along the [110] direction, so as to obtain "matches" on which pressure could be applied. The stress cell is described in Ref. 28. The magnitude of the pressure was measured using two strain gauges: the first was a piezoelectric quartz device located under the sample; the second was the splitting of the exciton freely propagating in the GaAs substrate,[29] measured by reflectivity. Both gauges gave results in close agreement. The values of the various transition energies were extracted from reflectance spectra using the procedure described in Ref. 30.

B. Experimental results

Figure 1 shows the evolution of the low-energy part of the reflectance spectrum for sample D under several magnitudes of the [110] uniaxial stress. Two structures—(a) and (b)—appear, corresponding, respectively, to the e_1^w-h_1^w and e_1^w-l_1^w excitonic transitions.[21] The notations used are the same as in Refs. 13, 21, and 25: e, h, and l refer to electrons, heavy holes, and light holes, respectively, the subscripts indicating the quantum number of the

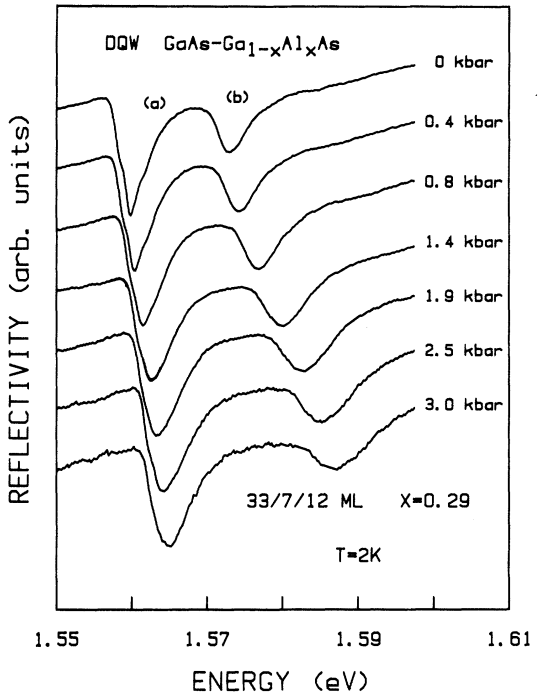

FIG. 1. Evolution of the low-energy part of the reflectance spectrum of sample D, as a function of the uniaxial stress applied along the [110] direction. The widths of the layers are L_1, h, and $L_2 = 33$, 7, and 12 monolayers, respectively.

electronic states and a superscript w or n indicating whether a given level originates from the wider well or from the narrower one. For instance, the first heavy-hole level of the narrow well might give rise to the third heavy-hole level of the whole DAQW; it is thus denoted h_3^n. The identification of the structures depicted in Fig. 1 is confirmed by their stress behavior. The heavy-hole exciton shifts towards high energies with a smaller slope than the light-hole exciton, as predicted by theoretical calculations.[13,29,31–33]

What is shown in Fig. 1 does not seem very different from what was previously obtained by optical spectroscopy on single quantum wells (SQW's).[34–36] However, a careful examination shows that the high-energy structure has a lower stress coefficient at high stress than at low stress. This effect will be addressed in the next section. The slight stress broadening of both structures is interpreted in terms of inhomogeneities of the strain over the sample.

Figure 2 presents the modifications induced by the stress on the high-energy part of the reflectance spectrum of sample D. At the top of the figure, the zero-stress spectrum exhibits five structures, labeled (a)–(e). One of them [structure (c)] is very weak and very difficult to follow, when stress is applied. The others are rather intense, and have previously[21] been identified as (a) e_2^n-h_2^w, (b) e_2^n-h_3^n, (d) e_3^w-h_2^w, and (e) e_2^n-l_2^n and/or e_3^w-h_3^n. These identifications were consistent with effective-mass calcu-

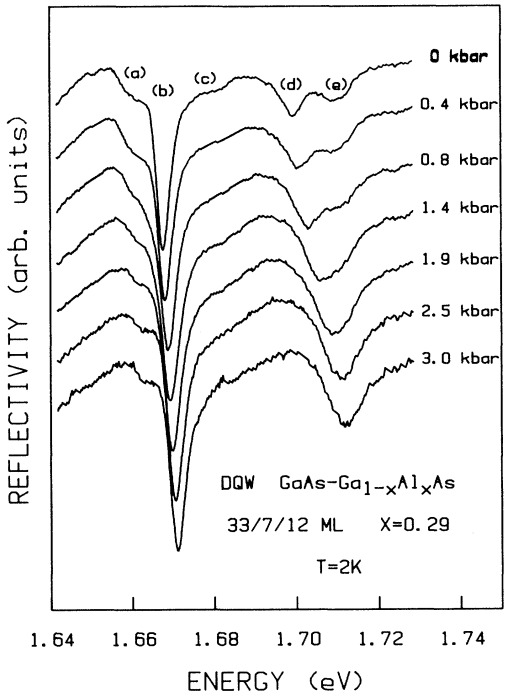

FIG. 2. Analog of Fig. 1, but for the high-energy range of the spectra of sample D. The general shape of the spectrum is strongly altered by the stress, due to excitonic features shifting at different rates.

lations, taking into account the usual material parameters and the following values of the respective layer thicknesses: $L_1=34$ monolayers (ML) for the wide well; $L_2=12$ ML, for the narrow well; and $h=8$ ML, for the intermediate barrier (1 ML = 2.83 Å). Reasonable values were estimated for the excitonic binding energies. The first result of our uniaxial-stress experiment is that our former interpretation of structures (d) and (e) was wrong. As a matter of fact, from Fig. 2, we see that line (d) corresponds to a light-hole exciton, since it shifts towards high energies faster than line (e), which overlaps the former, when the sample is stressed. The explanation for the doubt we could have, before performing stress experiments, is that we expected the observed optical transitions to be in the same order of energy as the theoretical ones. We thus took the above-mentioned values for the layer thicknesses and Rydberg energies. In fact, our present finding is rather compatible with the following values: $L_1=33$ ML, $L_2=12$ ML, and $h=7$ ML; then, structure (d) can be attributed to e_2^n-l_2^n and (e) to e_3^w-h_3^n and e_3^w-h_2^w, since these transitions are very close in energy. The binding energy of the light-hole exciton would then be ~ 10 meV, while it would be very small (similar to the Rydberg energy of bulk GaAs) for the two *fairly allowed* heavy-hole excitons. This interpretation is quite reasonable, if we compare it to simple calculations[16,18,24,25] of the Rydberg energies in DAQW's. Thus, the e_2^n-l_2^n transition would be pushed to an energy lower than e_3^w-h_3^n and e_3^w-h_2^w would, due only to a stronger Coulombic binding. This effect could not be predicted by a simple calculation of subband-to-subband transition energies. Moreover, as already noticed,[21] the agreement between theory and experiment is difficult to obtain, since we deal with a very intricate situation in which various transitions are superimposed on each other. This is partly due to the incidental proximity of the h_3^n and h_2^w levels, whose wave functions are delocalized over the whole DAQW, allowing the appearance of certain *interwell* excitons, such as e_2^n-h_2^w or e_3^w-h_3^n. The use of uniaxial stress allowed us to improve our identification.

The same experiments were performed on sample C, which is made up of less strongly coupled quantum wells ($h=15$ ML). Figure 3 displays the lower-energy part of the reflectivity spectrum, for sample C, at various values of the applied [110] stress. The behaviors are comparable to those quoted above for sample D, i.e., a faster shift for the e_1^w-l_1^w exciton than for e_1^w-h_1^w, at low stress. At high stress, e_1^w-l_1^w presents a kind of saturation, and shifts in a rather parallel fashion to e_1^w-h_1^w. The stress broadening of the structures is smaller than that previously observed. The high-energy part of the spectrum of sample C is shown in Fig. 4. Here, the previous identifications of the transitions seem to stand: (a) e_2^n-h_3^n, (b) e_3^w-h_2^w, (c) e_3^w-h_3^n, and (d) e_2^n-l_2^n. Structure (d) effectively shifts faster than the (a)–(c). A fifth, weak transition, (e), is very difficult to assign, but clearly involves heavy holes, since this structure disappears into structure (d) at high stress.

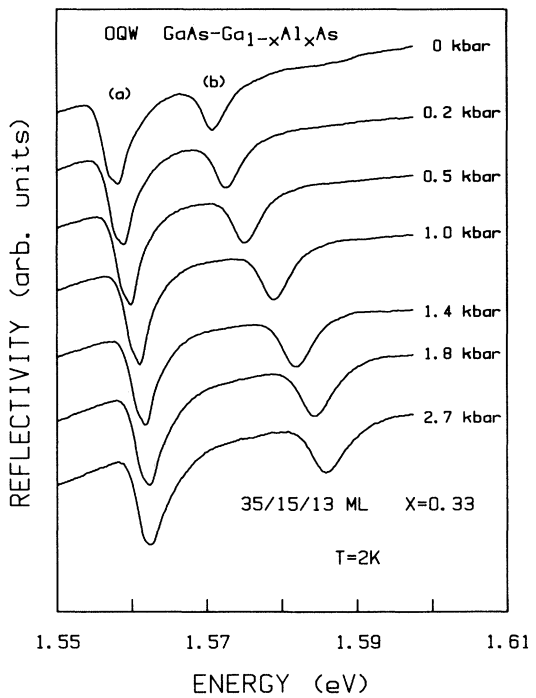

FIG. 3. Same as Fig. 1, for sample C. Structures (a) and (b) correspond, respectively, to the e_1^w-hh_1^w and e_1^w-lh_1^w excitons, as indicated by their respective stress shifts.

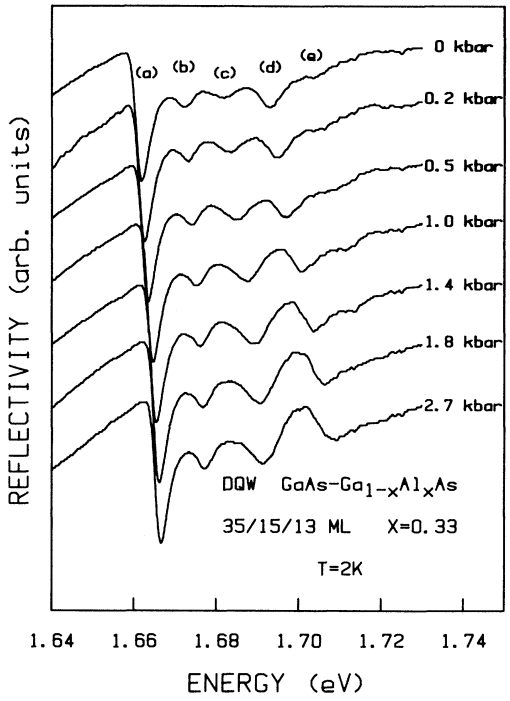

FIG. 4. High-energy zone of the reflectance spectrum for sample C, up to 2.7 kbar. The general aspect and its transformation under stress are completely different from the case of sample D, though the dimensions of the quantum wells are quite similar. This is an indication of the complexity of the system of excitonic states in DAQW's.

III. COMPARISON WITH THEORY: DISCUSSION

By using the effective-mass approximation, in the most general way, the μth wave function of an exciton in a quantum well can be written[37]:

$$\Psi^{\mu}_{\rm exc}(\mathbf{r}_e,\mathbf{r}_h)=\frac{\Omega}{\sqrt{A}}\sum_{m,m'} C^{m'}_{m} F^{\mu}_{m,m'}(\rho,z_e,z_h) \times\psi^{c}_{m}(\mathbf{r}_e)\mathbf{K}\psi^{v}_{m'}(\mathbf{r}_h)\ , \quad (1)$$

where Ω is the total volume of the system, A is the area of the quantum well, and ρ describes the in-plane relative motion of the electron-hole pair. The conduction- and valence-band Bloch waves at the direct band gap are ψ^{c}_{m} and ψ^{v}_{m}, respectively. The magnetic number m takes values of $\frac{1}{2}$ and $-\frac{1}{2}$, while m' runs from $-\frac{3}{2}$ to $+\frac{3}{2}$. $\mathbf{K}$ is the time-reversal operator, which transforms the Bloch function of the valence-band electron into that of a hole. μ represents the electron and hole subband indices.

Solving the problem of a $1s$ exciton in a quantum well, using the general wave function of Eq. (1), requires tedious calculations. The complexity can be reduced by making a few reasonable approximations.

(i) The eight-fold degeneracy of the $1s$ exciton is partly lifted by the electron-hole exchange interaction. As this quantity is very small, we can neglect it with satisfactory accuracy; this enables us to omit the electron label and to work with a 4×4 Hamiltonian matrix:

$$H_{\rm ex}=\begin{array}{c} \begin{matrix} |\frac{3}{2}\rangle & |\frac{1}{2}\rangle & |-\frac{1}{2}\rangle & |-\frac{3}{2}\rangle \end{matrix} \\ \begin{bmatrix} P_{11} & P_{12} & P_{13} & 0 \\ P^{*}_{12} & P_{22} & 0 & P_{13} \\ P^{*}_{13} & 0 & P_{22} & -P_{12} \\ 0 & P^{*}_{13} & -P^{*}_{12} & P_{11} \end{bmatrix} \end{array} + \left[H_e-\frac{2}{\varepsilon|\mathbf{r}_e-\mathbf{r}_h|}\right]\cdot\mathbb{1}_4\ , \quad (2)$$

where $\mathbb{1}_4$ is the 4×4 unit matrix.

(ii) We assume that, in these samples, the Coulomb interaction is small compared to the sum of the electron and hole confinement energies. Subsequently, it poorly influences the conduction- and valence-band envelope functions. This approximation, together with the neglect of $\mathbf{k}\cdot\mathbf{p}$ coupling, enables us to separate the conduction- and valence-band physics. Numerical values for the heavy- and light-hole exciton Rydberg energies will be computed within this context.

Thus, on one hand, the envelope functions for the conduction states are the eigenfunctions of the following Hamiltonian:

$$H_e=-\frac{\partial}{\partial z_e}\left[\frac{1}{m^{*}_{e}(z)}\frac{\partial}{\partial z_e}\right]+V_e(z_e,\sigma)\ , \quad (3)$$

$$V_e(z_e,\sigma)=V_e(z_e,0)+a_c(e_{xx}+e_{yy}+e_{zz})\ . \quad (4)$$

σ is the external stress, a_c is the deformation potential of the conduction band, and the e_{ij} are the components of the strain tensor.

On the other hand, the valence-band eigenstates are those of the left-hand 4×4 matrix whose elements P_{ij} can be decomposed into three parts:

$$P_{ij}=\mathcal{H}^{\rm LK}_{ij}+D_{ij}+V_H(z_H)\delta_{ij}\ . \quad (5)$$

$\mathcal{H}^{\rm LK}_{ij}$ is the Lüttinger-Kohn Hamiltonian,[38] D_{ij} is the Bir-Pikus term,[39] and $V_H(z_H)$ is the valence-band potential, which is taken at 33% of the total gap mismatch between GaAs and $Ga_{1-x}Al_xAs$, as is now well established.

The physics of quasibidimensional excitons has been considered under various approaches that differ from each other in the strength of their basic assumptions. To the best of our knowledge, the most accurate calculations are those proposed by Bauer and Ando[40] and by Andreani and Pasquarello,[41] who take into account a variety of physical effects, such as valence-band mixing, Coulomb coupling between excitons belonging to different subbands, and mismatch between the dielectric constants of the two host crystals. A few authors have examined the situation of exciton states in coupled double quantum wells, investigating both symmetrical[18,16] and asymmetrical[24] structures. Their models neglect, for simplicity, the intersubband couplings, which means that extra diagonal terms of Eq. (2) are neglected in order to work at $\mathbf{k}_{\perp}=0$, where the calculations are not too complicated. We extended the calculation to the exciton states associated with higher electron and hole subbands and

treated the problem variationally, describing the exciton by using the following ansatz:

$$F^{p,q}_{m,m'}(\rho,z_e,z_h)=(\sqrt{2}/\sqrt{\pi})\beta^{p,q}\cdot\chi^{p}_{m}(z_e)\cdot\chi^{q*}_{m'}(z_h)\times\exp(-\beta^{p,q}\rho)\ , \quad (6)$$

where μ of Eq. (1) is represented by the pair p,q of electron and hole subband indices, $\chi^{p,q}$ is the envelope function of the electron (hole), and $\beta^{p,q}$ is a variational parameter.

Figure 5 displays the change of the Rydberg energies computed for various widths of the intermediate barrier, in DAQW's having well widths similar to those of our samples. Note the collapses of some of the Rydberg energies when the vertical excitons tend to become oblique. This is never observed for excitons such as e_1^w-h_1^w and e_1^w-l_1^w, which remain vertical for any value of the internal-barrier width. This $\mathbf{k}_\perp=0$ approximation should lose some validity when the confined levels incidentally become very close in energy. In a previous work,[13] we presented a theoretical study of the electronic states in GaAs-(Ga,Al)As DAQW's subjected to in-plane uniaxial stress. We have demonstrated the possibility of obtaining a great diversity of *interwell* and *intrawell* anticrossings between the confined states, because of the lack of symmetry. In such situations, the Coulombic interaction may sensitively alter the stress-induced behavior of the optical transition energies and oscillator strengths, near the high-mixing points; a heavy multiband calculation is required, we believe, to obtain accurate values of the Rydberg energies. We have calculated the stress-induced shift of the optical transitions in our samples, making the assumption that the Rydberg energies are stress independent, even when valence-confined states become resonant. This was done by including the Rydberg energies calculated at zero stress, and at $\mathbf{k}_\perp=0$ in the 4×4 matrix of Eq. (2), and then calculating the optical transition energies for various values of the stress.

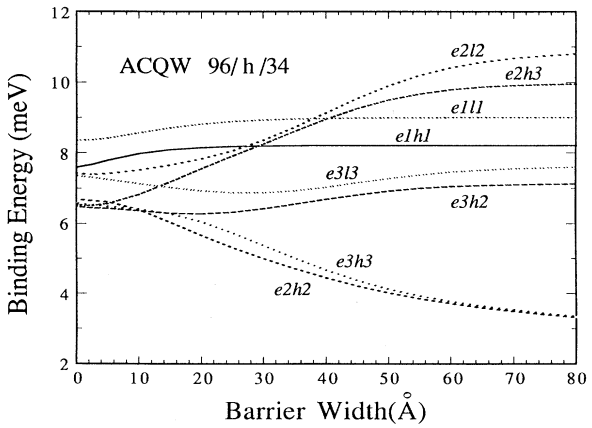

FIG. 5. Calculation of the binding energies of the various $1s$ exciton states that appear in a DAQW with well widths similar to those of our samples (96 and 34 Å) and with varying barrier thicknesses. A kind of anticrossing behavior between vertical and oblique excitons occurs around a barrier width of ~20 Å. These excitons involve the two quasiresonant h_2 and h_3 valence states, which are delocalized over the whole DAQW.

We wish to briefly remind the reader of a few predictions concerning DAQW's that can be extracted from Ref. 13: Due to the asymmetry of these structures, there are no longer any parity-related selection rules for optical transitions, and thus the possibility of observing *interwell transitions is enhanced.* Without the help of external perturbations, such as on-axis electric fields, these transitions are possible, as mentioned above, only for very particular designs of the DAQW's, for which levels originating from both wells incidently come very close to each other. Besides, in regard to the influence of uniaxial stress, peculiar effects are expected: *In the envelope-function approximation,* an interband crossing (without any mixing) between, say, an l_1 state and an h_2 state is possible in symmetrical structures, because of their opposite parities. This is no longer true in DAQW's. Models that include the exact symmetry of the problem[42] *always* predict an anticrossing of these states in *any* (001)-grown heterostructure between compounds that share either a common anion or a common cation. Upon comparison to the case of symmetrical microstructures, we believe that the asymmetrical nature of our samples will enhance such an anticrossing, when levels are pushed close to each other. This anticrossing should thus be observed more readily than in SQW's. Moreover, such anticrossings can involve valence states formerly mainly localized within the same well (*intrawell* anticrossings) or in separate wells (*interwell* anticrossings): The stress can produce a kind of resonant tunneling between light- and heavy-hole states, across the intermediate barrier. These peculiar properties are thus expected to have measurable repercussions on the stress-induced energy shifts of the optical transitions.

Let us now focus on the results of our stress experiments. The stress-induced energy shifts of the various observed excitonic transitions are compared, in Figs. 6 and 7, to the calculated behaviors of the corresponding transitions. In the low-energy range, involving states related to the wider well, the agreement between theoretical and experimental slopes is rather good. However, we remark that e_1^w-l_1^w exchanges its slope with e_1^w-h_2^w, for stresses higher than ~2.6 kbar, while no transition is visible at high energy. Such a behavior is not that previously observed by reflectance measurements on SQW's.[31] This unambiguously proves that a *strong anticrossing* occurs due to the asymmetry of the DAQW.

Similar behavior is observed for sample C (see Fig. 7), but the anticrossing occurs near 1.5 kbar. We calculated the influence of [110] stress on the valence-band structure of DAQW's in a previous paper.[13] Such a calculation was repeated for the present situation: The l_1^w-h_2^2 anticrossing is calculated near 2.2 kbar for both samples. This is not found experimentally, due to the excitonic interaction. A similar observation was reported by Ferreira *et al.*[22] They aligned the *electron* levels by biasing a DAQW of appropriate design, and found that the anticrossing occurred for a bias value that was not predicted by a band-to-band calculation, but could be accounted for if one included excitonic mixing as a perturbation.

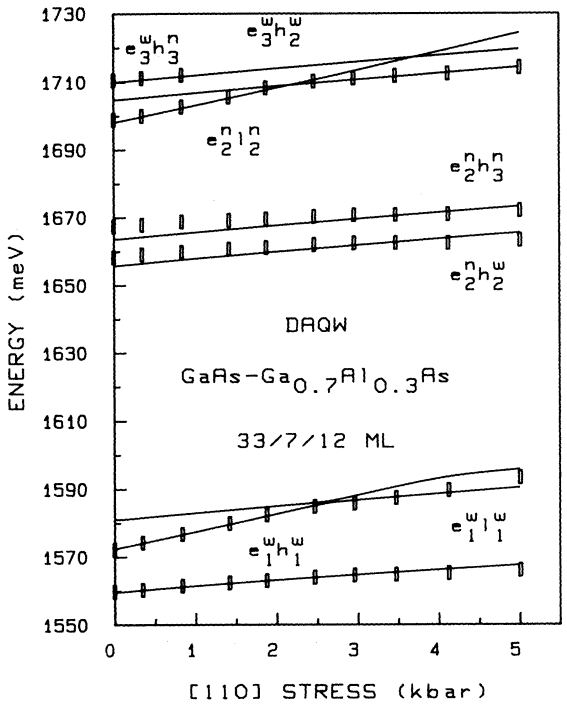

FIG. 6. Plot of the [110] stress-induced energy shift of the optical transitions in sample D. Open rectangles mark the position of the measured excitonic transitions. Solid lines represent the results of the calculation described in the text, ignoring any stress-induced mixing between excitonic states and any change in the binding energies.

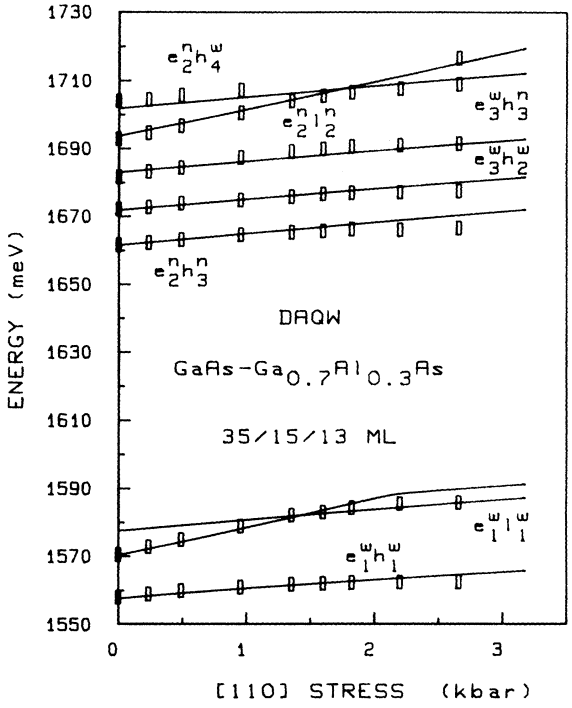

FIG. 7. Analog of Fig. 6, but for sample C. The calculated slopes of the transition energies are in close agreement with the measured ones, and the stress positions of the anticrossings are related to the excitonic nature of the transitions. These positions would be different if the corresponding band-to-band transition energies were plotted.

The exciton trial function of Ferreira *et al.* is comparable to that used in the present study, but here we deal with inter-valence-band couplings.

Inspection of the agreement between theory and experiment, in the high-energy range, provides confirmation of the importance of excitonic effects. Roughly comparing the order of magnitude of typical two-dimensional-excitonic binding energies (~ 10 meV) to the separation between two subbands, at an anticrossing point (sometimes less than 1 meV), one is easily convinced that a simple perturbational treatment is not appropriate. As shown in Fig. 6, the measured shifts of e_2^n-h_2^w and e_2^n-h_3^n [structures (a) and (b) of Fig. 2] are in good agreement with the calculations. The case of e_2^n-l_2^n and e_3^w-$h_2^w + e_3^w$-h_3^n transitions [structures (d) and (e) of Fig. 2] is much more interesting since it shows real stress-induced *excitonic mixing*. As explained above, the e_2^n-l_2^n transition turns out to be at lower energy than e_3^w-h_2^w, due only to a larger excitonic binding energy. Thus our calculation cannot predict the observed behavior: The light-hole exciton is repulsed by its high-energy companion, which provokes a bending of its stress shift. The e_3^w-$h_2^w + e_3^w$-h_3^n excitons [structure (e)] are no longer resolved, so that, in this range of applied stress, no evident increase of their slope is measured—we just observe a broadening of the experimental feature.

We have here an example of a situation in which a correct theoretical treatment of excitons in DAQW's under stress is required to analyze the experiment quantitatively. The situation is not so drastic for sample C, where the barrier is thicker: e_3^w-h_2^w and e_3^w-h_3^n excitons are well resolved, but the stress-induced coupling of vertical e_2^n-lh_2^n and oblique e_2^n-hh_4^W excitons is observed and leads to a broadening of the experimental features.

IV. CONCLUSIONS

It is well known that the application of uniaxial stress perpendicular to the growth axis of a quantum well provokes couplings between light- and heavy-hole states. We showed in a previous paper that this effect could produce interesting resonant-tunneling processes in asymmetrical double quantum wells. What is shown in the present article is that, in practice, optical spectroscopy reveals peculiar stress behaviors that seem characteristic of mixings between exciton states rather than between interaction-free carriers. This result is comparable to that obtained in a series of works by Ferreira *et al.*,[22,27] where the authors demonstrate the existence of resonances in the transfer of excitons between two biased quantum wells, rather than resonances between free-electron or hole levels.

In the present case of DAQW's, it seems, moreover, that a correct theoretical calculation of excitonic states

and energies is particularly tedious, due to the complexity of the couplings. This point is to be compared to the case of excitons in very wide single quantum wells, which should exhibit a variety of mixings, as predicted by Bauer and Ando.[40] However, the inclusion of all of the quoted physical effects in the treatment of excitons in DAQW's is particularly difficult; additional mathematical and numerical problems need to be resolved, in light of the case of single quantum wells.[43] This is out of the scope of the present experimental work.

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