RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 31, NUMBER 12 15 JUNE 1985

Optical investigation of a new type of valence-band configuration in $In_xGa_{1-x}As$-GaAs strained superlattices

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(Received 8 April 1985)

We present optical absorption experiments in strained $In_xGa_{1-x}As$-GaAs superlattices grown by molecular-beam epitaxy. From our data, we show that ground heavy- and light-hole states are confined in the $In_xGa_{1-x}As$ and in the GaAs layers, respectively. We also use a new method to observe the optical selection rules in such two-dimensional structures.

Among semiconductor superlattices, which are two-dimensional systems first proposed by Esaki and Tsu[1] in 1970, the GaAs-$Ga_xAl_{1-x}As$ structures have been the most thoroughly studied. This system is usually said to be of type I: Owing to the different band gaps of the host semiconductors, a superpotential is created in the z direction perpendicular to the plane of the layers and tends to confine both electrons and holes in the smaller gap material, i.e., in the GaAs layers.[2] On the other hand, in InAs-GaSb superlattices, the top of the GaSb valence band lies at higher energy than the bottom of the InAs conduction band and, as a result, electrons are mainly localized in the InAs layers while holes are confined in the GaSb ones:[3] Such a system, where electrons and holes are spatially separated, is said to be of type II. In this work, we show for the first time that an alternative situation can be reached in strained $In_xGa_{1-x}As$-GaAs multiquantum wells or superlattices. Indeed, because of the strain, which is due to the lattice mismatch between the involved semiconductors, such a structure is, at the same time, of type I for heavy holes and of type II for light holes.

The potential interest in these strained semiconductor heterostructures has already been demonstrated.[4-6] The possibility of getting rid of the drastic lattice-matching condition in the heteroepitaxy of sufficiently thin layers[7] enlarges the choice of materials that can be grown on a given substrate. In the considered system, for an In content of 15%, the ternary layer thickness should be smaller than 120 Å to obtain good-quality layers where the lattice mismatch is totally accommodated by elastic strain. In this elastic limit, in an $In_xGa_{1-x}As$/GaAs heterostructure grown on a GaAs substrate, only the $In_xGa_{1-x}As$ layers are strained, and the strain amplitude is independent of their thicknesses. Accordingly, the valence-band degeneracy should be lifted. In such a system, the relative positions of the bands in the $In_xGa_{1-x}As$ wells and the GaAs barriers can lead to several plausible configurations of the quantizing superpotentials, which are shown in Fig. 1. By analyzing optical data obtained on a series of $In_xGa_{1-x}As$/GaAs multi-quantum-well structures with $x \simeq 0.15$, we show that, around this composition, the GaAs valence band actually lies between the $In_xGa_{1-x}As$ heavy- and light-hole bands.

Figure 2 shows absorption spectra obtained at 77 K on three samples consisting of 10 periods of alternate layers of GaAs (thickness 200 Å) and $In_xGa_{1-x}As$ (thickness 50, 100, 120 Å, $x \simeq 0.15$) grown by molecular-beam epitaxy on a (100) GaAs substrate. They have steplike shapes and exhibit definite excitonic structures, which are also observed at 300 K. This reveals the high optical quality of the samples and shows that we are actually dealing with two-dimensional systems. Moreover, x-ray double-diffraction-spectra analyses[8] indicate that, in samples A and B, the $In_xGa_{1-x}As$ layers completely accommodate the 1% lattice mismatch by elastic strain, and transmission-electron-microscopy analyses demonstrate that these structures are almost dislocation free. We will first discuss the results obtained on these two samples. Because they are well aligned with the substrate, and as we know quantitatively the stresses and the In content in these ternary layers from x-ray measurements, we can calculate the relevant band extrema in the strained $In_xGa_{1-x}As$. They can be viewed as superlattices built from (i) small-gap $In_xGa_{1-x}As$ layers under compressive biaxial (in-plane) stress and (ii) unstrained GaAs layers. The stress applied on the $In_xGa_{1-x}As$ layers is equivalent to the sum of a hydrostatic pressure and a tensile uniaxial stress along the z direction, leading to the following effects: (i) an increase of the band gap and (ii) a splitting of the valence band, the heavy-hole band lying at a higher energy than the light-hole one[9] (Fig. 1). Table I shows the characteristics obtained for the three samples. Sample C will be discussed later. The unstrained ternary band-gap variations versus the In content (around 15%)

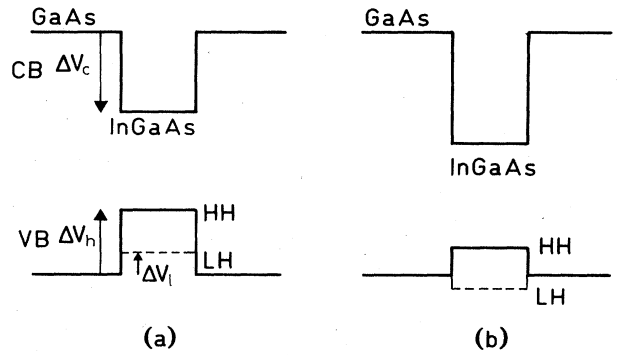

FIG. 1. Possible energy-band configurations in a strained InGaAs-GaAs quantum well; CB and VB refer, respectively, to the conduction and valence bands of the host materials. In the InGaAs layers, the valence band is split into heavy- (HH) and light- (LH) hole bands as a result of the strain arising from the lattice mismatch between the two materials.

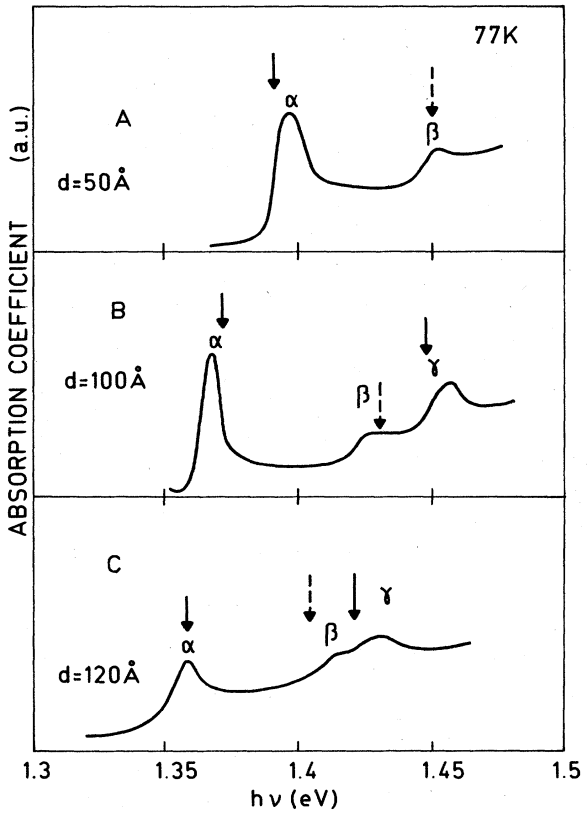

FIG. 2. Absorption spectra at 77 K in three samples with different $In_xGa_{1-x}As$ layer thicknesses d. The arrows indicate the calculated energies for the transitions associated with heavy ($\rightarrow$) and light ($- - \rightarrow$) holes.

have been calibrated by 77-K absorption experiments on 2-μm-thick samples. The In composition x in these reference samples has been measured by microprobe and they were then shown to be unstrained by x-ray double diffraction. The results are very close to those reported by Leu *et al.*,[10] i.e.,

$$E_g(x) = 1.508 - 1.47x + 0.375x^2 \ .$$

The elastic constants and the deformation potentials of the alloy are taken from a linear interpolation of the values for the binaries. We use $a = -8.4$ eV (Ref. 11) and $b = -2$ eV (Ref. 9) for GaAs and $a = -6$ eV (Ref. 12), $b = -1.8$ eV (Ref. 13) for InAs. In the calculation of the valence-band splittings given in Table I, we include the effect of the coupling between the light holes and split-off bands. This effect reduces the splitting of the heavy and higher-energy light-particle valence bands by 8–10 meV. We can also note that the resulting superlattice potential has the same symmetry as the strain, so that, in an envelope-function approximation,[14] one can still use $|\frac{3}{2}, \pm\frac{3}{2}\rangle z$, $|\frac{3}{2}, \pm\frac{1}{2}\rangle z$, $|\frac{1}{2}, \pm\frac{1}{2}\rangle z$ functions to calculate the eigenstates of the system. Moreover, the heavy-hole band is decoupled from the other bands in the z direction. For small stresses, the stress Hamiltonian is diagonal in this basis and the calculation differs from the $GaAs/Ga_xAl_{1-x}As$ one only by the fact that, due to the valence-band splitting in one of the constitutive materials, the heavy- and light-hole motions are now quantized along z by different potentials (Fig. 1). To determine the energies of the quantized levels of interest, only one unknown parameter remains, i.e., the conduction-band discontinuity at each interface, ΔV_c. In the following, we deduce from the experimental results a value for this parameter.

TABLE I. Sample characteristics: samples A and B are well strained onto the substrate, whereas for sample C, some stress relaxation has occurred. E_{gh} (E_{gl}) would be the heavy-hole (light-hole) band gap in a bulk $In_xGa_{1-x}As$, under the same strain conditions as in the superlattice.

	d (Å)	x	E_g (eV) (unstrained)	E_{gh} (eV)	$E_{gh} - E_{gl}$ (eV)
A	50	0.17	1.268	1.325	0.079
B	100	0.15	1.297	1.348	0.071
C	120	0.15	1.297	1.34	0.062

In the absorption spectra, the lowest energy peak α corresponds necessarily to excitons involving the first electronic level $1e_1$ and the first heavy-hole level hh_1. The observation of excitonic absorption peaks indicates that electrons and heavy holes are both confined in the same material.[2] For sample A, where the gap difference between GaAs and strained $In_xGa_{1-x}As$ is $\Delta E_g = 185$ meV, this means that

$$0 < \Delta V_c < \Delta E_g \ ,$$

so that

$$0 < \Delta V_h < \Delta E_g \ ,$$

where ΔV_h is the heavy-hole band discontinuity between the two materials (Fig. 1). A simple quantum-well calculation shows that there can then be only one hh_n-e_n transition in sample A. The second transition β is then likely to be related to the first light-hole level lh_1. For sample B, the α and β transitions are attributed to excitons involving e_1, hh_1, and e_1, lh_1, respectively, and the γ one, similar in shape to the α one, is thought to correspond to excitons involving the second-electron (e_2) and heavy-hole (hh_2) levels.

We have checked this assignment by an on-edge excitation of the photoluminescence experiment, whose operation is shown schematically in the inset of Fig. 3. In our samples the GaAs substrate is transparent in the wavelength range of interest. If we focus a dye laser beam on a cleaved edge, because of the high refraction index of GaAs, the refracted beam inside the substrate is at a grazing incidence with respect to the plane of the layers and because the difference of the refractive indices in $In_{0.15}Ga_{0.85}As$ and GaAs is small [it can be estimated to be smaller than 3×10^{-3} from measured values of the refractive indices in $In_{0.53}Ga_{0.47}As$ and GaAs (Ref. 15)], the beam penetrates the superlattice with negligible refraction. This allows us to excite the superlattice luminescence with light which can be linearly polarized either along or perpendicular to z. Figure 3 shows excitation spectra obtained in sample B. The standard surface-excitation spectrum is consistent with the absorption data of Fig. 2. The on-edge perpendicular polarization spectrum (Fig. 3) is similar to it, because in a surface-excitation spectrum the light is also mainly polarized in the plane of the layers inside the sample. On the other hand, when the incident laser light is polarized along z, the heavy-hole-related transitions are forbidden, while the light-hole ones are al-

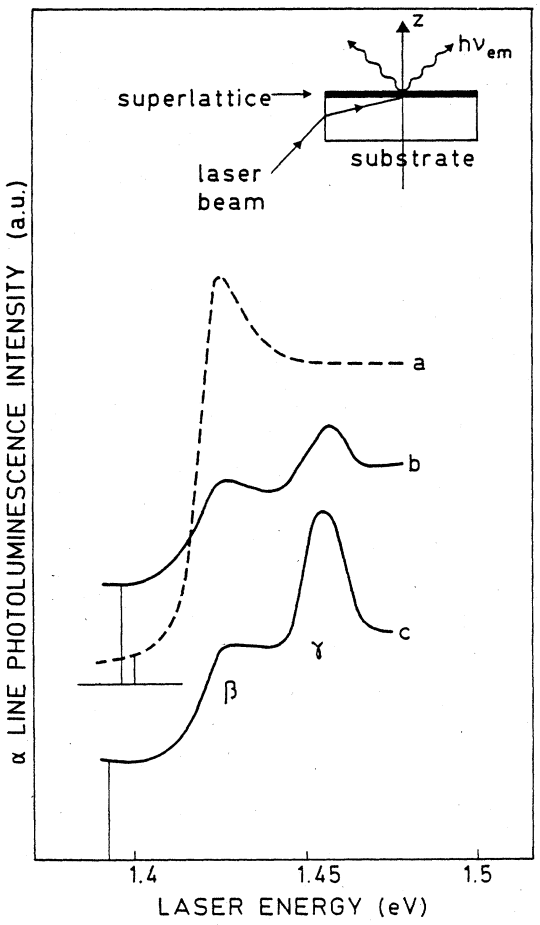

FIG. 3. On-edge excitation spectra obtained in sample B for light polarizations parallel (a) and perpendicular (b) to the z axis. The experimental setup is shown in the inset. (c) Excitation spectrum recorded in the standard configuration, where the laser beam is focused on the sample surface.

lowed,[9] and we do not observe the 1.46-eV γ transition, nor the absorption due to the ground electron, heavy-hole continuum states. However, the 1.43-eV β transition is still observed, and this supports our previous interpretation.

In order to estimate $\Delta V_c(x)$, we have expressed the conduction-band discontinuity as a percentage of the band-gap difference between GaAs and $In_xGa_{1-x}As$ under the hydrostatic part of the strain. The energies of the five transitions observed in samples A and B have been fitted with only this one adjustable parameter. We have used an envelope-function model[14] for the superlattice and included the stress effects. The excitonic binding energies have been taken to be equal to 8 meV, which would be correct in this well-thickness range.[16] The best agreement has been found to correspond to the situation shown in Fig. 1(b), and gives

$$\Delta V_c = 126 \text{ meV}, \quad \Delta V_h = 60 \text{ meV}, \quad \Delta V_l = -19 \text{ meV}$$

for sample A, and

$$\Delta V_c = 110 \text{ meV}, \quad \Delta V_h = 52 \text{ meV}, \quad \Delta V_l = -18 \text{ meV}$$

for sample B. In this configuration, the GaAs layers (200 Å thick) become the wells, and the $In_xGa_{1-x}As$ layers the barriers for the light holes. The width of the GaAs wells leads to a small confinement energy of 3 meV for lh_1. The $In_xGa_{1-x}As$ barriers are 20 meV high so that the light-hole envelope functions are leaking in these layers. Furthermore, for $In_xGa_{1-x}As$ layers of small thicknesses, they are strongly overlapping with the e_1 electron wave functions, so we can observe the $lh_1 - e_1$ transition. For larger $In_xGa_{1-x}As$ layer thicknesses, this transition should become weaker, and the transition between the light-hole band edge (LH) in $In_xGa_{1-x}As$ and e_1 should be observed.

We want to point out that in this configuration the energy position of the $lh_1 - e_1$ transition is very sensitive to the conduction-band offset because the lh_1 position scarcely depends on it, as it stays close to the GaAs valence-band extremum. In the limit between the two possible configurations [Figs. 1(a) and 1(b)], when the light-hole band extrema in GaAs and $In_xGa_{1-x}As$ are at the same energy, the β transition in sample B should be found 2 meV higher in energy than the γ one which should appear at 1.442 eV. The fact that $hh_2 - e_2$ appears at a higher energy than $lh_1 - e_1$ in sample B indicates that the actual valence-band configuration is that of Fig. 1(b).

Let us note that the value of the conduction-band offset (ΔV_c) could have been obtained by simple considerations of the number of levels in sample B and on the energy difference between the α and β transitions in sample A (see the Appendix).

The case of sample C is particularly interesting because the x-ray double-diffraction spectrum cannot be fitted exactly with the model described in Ref. 8 and there is thus a certain inaccuracy in the determination of the composition of the InGaAs layers. If we assume the superlattice to be well aligned with the substrate (perpendicular parameter remains the same as that of GaAs), the fit of the x-ray double-diffraction spectrum would lead to $x = 13\%$. However, in this case, the calculated strained heavy-hole band gap (E_{gh}) would be 1.37 eV which is already 10 meV higher in energy than the observed α absorption peak which has the shape of an excitonic peak and corresponds obviously to the hh_1-e_1 transition. Owing to the confinement energies $hh_1 - e_1$ should be 23 meV higher than E_{gh}. We are thus led to make the hypothesis that the superlattice has relaxed as a whole with respect to the substrate. It is indeed possible to find a value of x which fits the x-ray-diffraction profile and agrees with the $hh_1 - e_1$ transition energy deduced from the position of the α absorption peak. We take for the condition-band discontinuity ΔE_c the same percentage compared with the band-gap difference ΔE_G as in the case of samples A or B. The results are given in Table I. The In composition is found to be close to 15% and the superlattice in-plane parameter is $0.35\bar{a} + 0.65 a_{GaAs}$ where $\bar{a}$ would be the in-plane parameter of the superlattice if it was completely relaxed from the substrate. The calculated energies of the two other transitions β and γ are shown in Fig. 2. The agreement is not perfect and shows that the strain relaxation in this sample is perhaps more complicated. We would, however, insist on the fact that in the case of partly relaxed strained superlattices, x-ray diffraction is not sufficient to determine the composition and the thickness of the InGaAs layers and that optical measurements give much more precise data.

The authors wish to thank G. Le Roux who performed the x-ray measurements, P. Henoc for the transmission-electron-microscopy analysis, and G. Bastard, P. Voisin, and

M. Voos for fruitful comments and discussions. Laboratorie de Bagneux is a Laboratoire associé au Centre National de la Recherche Scientique (No. 250).

APPENDIX

It is possible to obtain the value of the conduction-band offset (ΔV_c) by a simple analysis of the number of levels in sample B and on the distance between the α and β transitions in sample A. The starting point is that the band-gap difference between the well and the barrier is $\Delta E_{gh}=185$ meV for the heavy-hole band and $\Delta E_{gl}=92$ meV for the light-hole valence band. In sample B, there are at least two electronic levels, so that $\Delta V_c > 62$ meV and $\Delta V_l < 30$ meV. If ΔV_c depends linearly on the composition, in sample A, $\Delta V_l < 36$ meV. If we are in the configuration of Fig. 1(a), the heavy- and light-hole exciton binding energies should be nearly equal, so that

$$E(\gamma)-E(\alpha)=\Delta V_h-\Delta V_l+lh_1-hh_1$$

if

$$\Delta V_h=36 \text{ meV}, \quad lh_1=25 \text{ meV} ,$$

so that $lh_1-\Delta V_l \simeq 11$ meV. As soon as $\Delta V_h > 35$ meV, then $hh_1 \simeq 15$ meV; this, together with the experimental value of $E(\gamma)-E(\alpha)=55$ meV, leads to $\Delta V_h \simeq 60$ meV, which is smaller than the valence-band splitting.

If we are in the configuration of Fig. 1(b), the light-hole exciton, if it exists, should have a very small binding energy, so that

$$E(\gamma)-E(\alpha)=\Delta V_h+lh_1-hh_1+E_b ,$$

where E_b is the binding energy of the heavy-hole exciton. With $E_b=8$ meV, $hh_1=15$ meV, and $lh_1=3$ meV, $\Delta V_h \simeq 59$ meV.

In both cases, we obtain the same estimate, which is only coherent in the second case.

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