Optical properties of $GaSb/Al_{0.4}Ga_{0.6}Sb$ multiple quantum wells

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The optical properties of $GaSb/Al_{0.4}Ga_{0.6}Sb$ multiple quantum well structures grown by molecular beam epitaxy on GaAs substrates were studied through absorption and photoluminescence measurements. Despite the various difficulties in growing good quality GaSb-based heterostructures a clearly resolved splitting between the absorption peaks related to the $n=1$ heavy and light hole excitons was observed in the energy range 0.85–1.15 eV even at room temperature. A detailed line shape analysis of the excitonic absorption peaks has been carried out in the GaSb/(AlGa)Sb MQW system: the reliability of such analysis was confirmed by the fact that the experimentally determined ratio between heavy to light ''in-plane'' reduced masses is in excellent agreement with the ratio in the literature. The energies of the interband electronic transitions, calculated within the envelope function approximation including strain effects, show a discrepancy with experiments interpreted in terms of interfacial roughness. The values of band offset and nonparabolicity coefficients resulting from the analysis are in good agreement with the data reported in the literature.

I. INTRODUCTION

Increasing attention has been recently devoted to GaSb-based quantum wells (QW) and superlattices[1–3] because the band gap of GaSb and related ternary and quaternary compounds matches a wavelength region of technological importance for optical communication system; moreover, quantum well infrared photodetectors, operating for normal incidence intersubband absorption,[2,4,5] and infrared modulators, taking advantage of the quantum confined Stark effect,[3,6] have been projected.

The realization of GaSb based structures of good quality is affected by some difficulties: (i) a relatively high density of background native acceptors;[7] (ii) surface stability during the growth; (iii) a lattice mismatch between the GaSb and AlSb (estimated to be 0.65%), significantly higher than the nearly perfect lattice match obtainable in the GaAs/(AlGa)As system: interfaces of lower quality are therefore expected in the antimony based MQWs. In addition, in view of their possible integration into optical devices, the characterization of GaSb/AlGaSb structures grown on GaAs substrates becomes helpful: the main limit to the crystallographic quality of such MQWs is given by the effects of strain relaxation at the interface between GaAs and the antimonide compound due to the significant lattice mismatch ($\approx 7\%$) between them, which induces a high density of threading dislocations (10^7 cm^{-2}; see Ref. 8).

The study of the optical properties of MQWs allows for a satisfactory investigation of the electronic transitions between confined states as a function of the structural parameters and permits a qualitative characterization of the interfaces, becoming a useful tool to optimize the growth conditions. This investigation has been exhaustively performed for long time in GaAs and InP based MQW systems; differently, at our knowledge only few authors searched into the optical properties of nominally undoped GaSb/AlGaSb MQWs through photoluminescence,[9–15] transmission,[12,13,16] reflectance,[17–20] or photocurrent[21] spectroscopy: generally the possibility to perform a complete analysis of the spectra, including the line shape analysis of the peaks, resulted considerably limited by the aforementioned difficulties affecting the growth of the antimonide based compounds.

In this work we report an accurate optical investigation of $GaSb/Al_{0.4}Ga_{0.6}Sb$ multiple quantum wells (MQW) of different well thickness: the samples were grown on GaAs substrates by stabilizing the growth temperature through different methods. Transmission and photoluminescence measurements were performed in the energy interval from 0.7 to 1.3 eV at different temperatures. The possibility of a reliable line shape analysis of the absorption peaks is also proved. The energies of the interband electronic transitions between quantum states are compared with the results of a calculation based on the envelope function approximation: the best agreement between experimental and theoretical data was obtained for values of the band offset and nonparabolicity coefficients which are in very good agreement with those reported in the literature; however, an uneliminable shift between the measured energies and the expectations of the model was found: such a discrepancy is explained in terms of interfacial roughness.

II. EXPERIMENTAL DETAILS

Nominally undoped (p-type[7]) $GaSb/Al_{0.4}Ga_{0.6}Sb$ MQW's were grown by molecular beam epitaxy (MBE) in an Intevac Gen II modular chamber, on [100] oriented semi-insulating (SI) GaAs substrate. The composition was measured by reflection high-energy electron diffraction (RHEED) oscillation periods with an accuracy within $\pm 5\%$. To check different growth conditions the samples were grown either In-bonded or In-free, and the growth temperature T_c was monitored through an optical pirometer. In order to avoid temperature variation during the In-free growth of a MQW due to IR absorption, a 2 μm thick GaSb absorbing buffer layer was grown beforehand. In the C376 sample (see Table I), the absorbing layer was interposed between MQW and substrate.[22] In the C405 sample the absorbing layer was grown on the *back* surface of the substrate and capped by a 1 μm thick GaAs layer to prevent its degradation during the growth of the MQW on the front surface of the substrate; the

TABLE I. Details of the structure of the samples investigated (n. C376, C405, C461, C462). Before the structure C405 was grown, a 2 μm thick GaSb layer was grown on the back of the sample and capped by a 1 μm thick GaAs layer, as explained in the text. x_b is the AlSb molar fraction of the barriers in the MQW.

		c376	c405	c461	c462
GaSb cap	thickness (Å)	100	100	99.3	100
	N periods	50	15	30	15
MQW	GaSb well (Å)	44.8	71	49.7	60
	(AlGa)Sb barrier (Å)	153.7	120	120.6	120
	x_b	0.42	0.403	0.395	0.392
	N periods		15		15
MQW	GaSb well (Å)		30		40
	(AlGa)Sb barrier (Å)		120		120
	x_b		0.403		0.392
Buffer	thickness (μm)	2	1	0.988	1
	AlSb molar fraction	GaSb	0.403	0.395	0.392
Substrate	type	GaAs	GaAs	GaAs	GaAs
	holding	In-free	In-free	In-bonded	In-bonded

absorbing layer was successively removed beforehand to perform the optical measurements. The latter method shows all the advantages of an In-free growth[22] avoiding, at the same time, the presence of a thick absorbing layers, which is detrimental for transmission measurements. Auger spectra taken at the growth surface of substrates on whose back side the absorbing GaSb layer was deposited resulted equivalent to the spectra taken at the growth surface of standard substrates immediately after the oxide desorption in the growth chamber.

The modeling of the structures was done with the aim to obtain a suitable optical density without exceeding the critical thickness for the strain relaxation t_s:[23] each sample included one or two series of MWQ, containing minimum 15 periods, grown pseudomorphic to an $Al_{0.4}Ga_{0.6}Sb$ (or GaSb) buffer layer. To improve the cristalline quality, in all the samples an $Al_{0.4}Ga_{0.6}Sb$ buffer layer 1 μm thick was interposed between the MQW and the substrate, except for the C376, where the GaSb absorbing layer guarantees the same result. The well width L_z was varied in the range 30–71 Å, just around the critical thickness for the confinement-induced Γ-L crossover $t_c \approx 40$–50 Å [depending on the barrier height,[9–11,16] in agreement with the theoretical value $t_c = 43$ Å predicted in GaSb/AlSb MQW (Ref. 24)]. Table I reports the details of the structures investigated.

All the samples were grown at $T_c = 550\,^{\circ}$C, a temperature used also by other authors,[9,10,13,14,17] with a V/III BEP ratio in the range 7–10; these conditions were optimized by maximizing the PL quantum efficiency of QW's grown on GaSb substrates. However, it is worth noting that a preliminary electrical characterization of similar nominally undoped MQW's, grown under the same conditions on (SI)GaAs without the thick AlGaSb (or GaSb) buffer layer, shows a surprisingly high *p*-type conductivity: under the hypothesis of an uniform distribution of carriers into the wells, the RT hole density approaches 10^{18} cm^{-3}, higher than the measured residual acceptors density in GaSb[25] and $Al_{0.4}Ga_{0.6}Sb$ single layers grown at the same temperature and BEP ratio. The details of the ''in-plane'' electrical investigation will be discussed in a forthcoming paper.

Transmission measurements at different temperatures (10–300 K) and low-T photoluminescence measurements were taken for all the samples in the spectral range of the electronic transitions (0.7–1.3 eV); in some selected cases PL spectra were also taken as a function of the temperature (10–300 K) and of the light power density (to 25 W/cm^2 on the sample). The absorption coefficient was obtained from the transmission spectrum according to the standard approach.[26]

III. RESULTS AND DISCUSSION

Transmission and PL spectra of comparable quality were obtained for the different samples, except for the C376 sample, owing to the presence of the absorbing GaSb buffer layer. As example, Fig. 1 shows the set of absorption spectra taken at different temperatures for the sample C461: the hh_1-e_1 and lh_1-e_1 excitonic resonance peaks are evidenced by arrows; they are related to the direct transitions between the $n=1$ heavy (hh) or light (lh) hole and the $n=1$ electron quantum level. Excitonic transitions of order $n=2$ were here not revealed. The energy position of the peaks, $E_{hh,1}$ and $E_{lh,1}$, estimated within ± 1 meV, shifts vs temperature following the energy gap of GaSb, as expected (see the inset of the figure). The features of the spectra are very well resolved compared to other absorption data reported in the literature for GaSb/AlSb MQW's (Refs. 12,13, and 16) and are evident till to room temperature: the good quality of the spectra is confirmed by the possibility to perform on them a reliable line shape analysis, following the approach of Chemla *et al.*[29] The decoupled contributions to the $T=10$ K absorption spectrum for the same sample of Fig. 1 are shown in Fig. 2. Such analysis was performed by requiring that the integrated intensity of each peak was temperature independent (within the 15% of fluctuation), as expected for weak absorption, and the widths of the hh_1-e_1 and lh_1-e_1 peaks were comparable (differences within 0.5 meV).

To test the reliability of the analysis we verify the theoretical equality $A_{hh}/A_{lh} = 3(\mu_{hh}/\mu_{lh})^2$, where A_{hh} and A_{lh} are the integrated intensities of the hh_1-e_1 and lh_1-e_1 peaks, respectively, and μ_{hh} and μ_{lh} are the ''in-plane'' reduced mass of the two excitons, in the same order.[30] The value

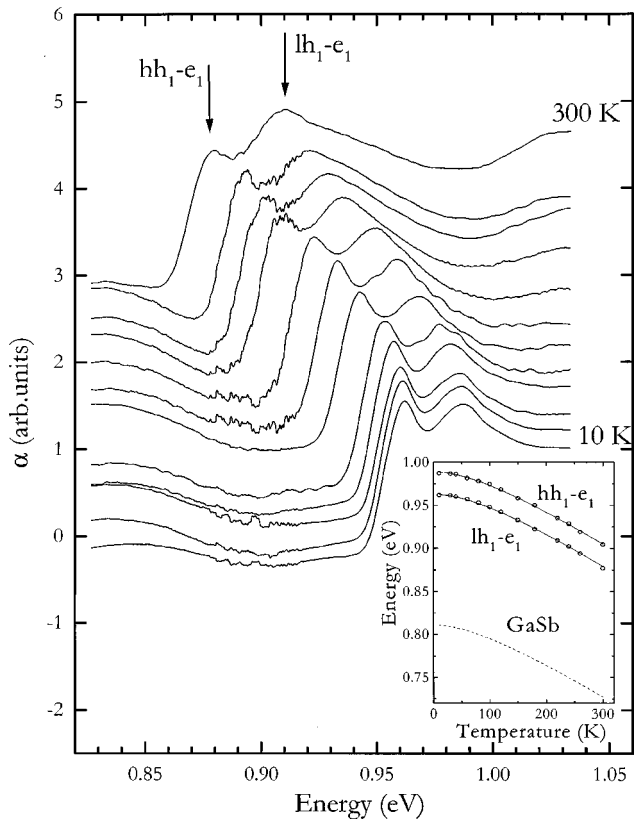

FIG. 1. MQW C461 ($L_z=50$ Å): absorption spectra at different temperatures between $T=10$ K and $T=300$ K ($T=10,30,40,60,80,120,150,180,220,240,260,300$). The hh_1-e_1 and lh_1-e_1 excitonic resonance peaks are indicated by arrows; the spectra are shifted along the y direction for clarity. The spectral resolution of the optical system was 0.8 meV. Inset: temperature dependence of the hh_1-e_1 and lh_1-e_1 transition energies for the same MQW in comparison with the temperature dependence of the energy gap of bulk GaSb (Ref. 27). The continuous lines are the best fits of the data given by the Varshni model [$\alpha=4.53\times10^{-4}$ eV/K and $\beta=186$ K for all the samples, equal to the parameters estimated for bulk GaSb by Ghezzi *et al.* (Refs. 27 and 28)].

$A_{hh}/A_{lh}=2\pm0.2$, resulting from the analysis, is in very good agreement with the calculated one of 1.92. The latter value was obtained by taking the electron effective mass equal to $m_c=0.0412$ and the ''in-plane'' masses of heavy and light holes equal to $m_{//}=1/(\gamma_1\pm\gamma_2)$, ($+$ for heavy holes and $-$ for light holes), where $\gamma_1=12.73$ and $\gamma_2=4.09$ are the Luttinger parameters for GaSb.[31,32] The decoupling procedure let us to confirm the energy positions of the peaks obtained directly from the transmission spectra within 2 meV; the fit of the dependence on temperature of the full width at half maximum (FWHM) for the hh_1-e_1 peak through the usual Chemla law[29] (see Fig. 2) gave a inhomogeneous and homogeneous broadening factor $\Gamma_o^{\rm abs}=19\pm1$ meV and $\Gamma_{\rm ph}=9.42$ meV (accuracy: $\pm5\%$) respectively. $\Gamma_{\rm ph}$ results to be comparable with data relative to bulk GaAs [of ≈7 meV (Ref. 33)] and to other MQW systems (see, e.g., Ref. 34); the high value of the inhomogeneous broadening will be briefly commented later.

In Fig. 3(a) the photoluminescence spectra taken at $T=10$ K for four MQW's are reported; no PL signal was revealed for the MQW's having 30 Å and 40 Å thick,[10] both embedded between the substrate and a second series of MQW of higher well width ($L_z>40$ Å). The PL peaks obtained for the different MQW's are slightly asymmetric, have comparable FWHM and are situated 3–15 meV below the corresponding hh_1-e_1 absorption peaks: the difference between PL and absorption peaks is generally attributed to a Stokes shift. In our case the fairly high density of background acceptors (resulting from the aforementioned electrical data) could suggest a relevant contribution due to transitions involving acceptor levels.[12] However, by varying the power density of the incident light between 0.25 to 25 W/cm^2 the PL spectrum shows a very weak shift to higher energies of about 4 meV and the peak intensity increases sublinearly, supporting the hypothesis of a dominant Stokes contribution.

As shown in Fig. 3(b) for the MQW C461, the energy position of the PL peak shifts to lower values as the temperature rises and slightly approaches the hh_1-e_1 absorption peak energy, although at the same time the intensity of the PL peak sharply decreases, reducing the reliability of this comparison. This clearly observable thermal quenching of the intensity is consistent with a thermally activated transfer of electrons from the Γ to L electron $n=1$ subband. The analysis of our data, by following Ru *et al.*,[35] gave an activation energy $E_a=19$ meV, roughly comparable with the expected separation between the Γ and L quantum states for a ≈50 Å thick QW (see, e.g., Ref. 9).

The experimental energy transitions were compared with the results of a calculation performed within the envelope function approximation for a single finite rectangular well.[36] In model were included non-parabolicity effects and band coupling through energy dependent effective masses for electron and light holes, defined in terms of the Luttinger parameters. The effects of strain were also estimated[23,37] by properly correcting the gaps and band offset values and including the thermal[38] and residual strain,[39] but ignoring the effect of strain on the effective masses.

The values of the band parameters assumed in our analysis are given in Table II: where not directly specified, the data relating to the $x=0.4$ barrier composition were taken as linear interpolation between those of GaSb and AlSb. The data of Alibert *et al.*[43] were considered for the fundamental gap at $T=0$ K of GaSb and $Al_{0.4}Ga_{0.6}Sb$. The VB discontinuity between these two compounds resulted equal to $\Delta E_{VB}=151\pm20$ meV and the non-parabolicity coefficients for the CB and the light hole valence band equal to $E_{oc}=0.989$ eV and $E_{o,lh}=0.504$ eV, respectively, obtained from the best fit of our data. These values agrees very well with similar data reported in the literature.[9,16,19,20,27,44]

The experimental data were fitted by considering the energies of the excited transitions (i) extrapolated at $T=0$ through the Varshni law, and (ii) roughly corrected for the exciton binding energy. The latter parameter was assumed to be $B_{hh}=B_{lh}=5.5\pm1$ meV, both for heavy and light hole excitons, by following the calculation of Giugno *et al.*[45] and by taking into account the x dependence of the binding energy and the small difference between B_{hh} and B_{lh}:[46] the variation of B in the spectral region of our interest[45] was neglected, it being comparable with the accuracy of the calculation, which does not include effects such as band nonparabolicity and subband coupling,[47] or correction for strain.[48]

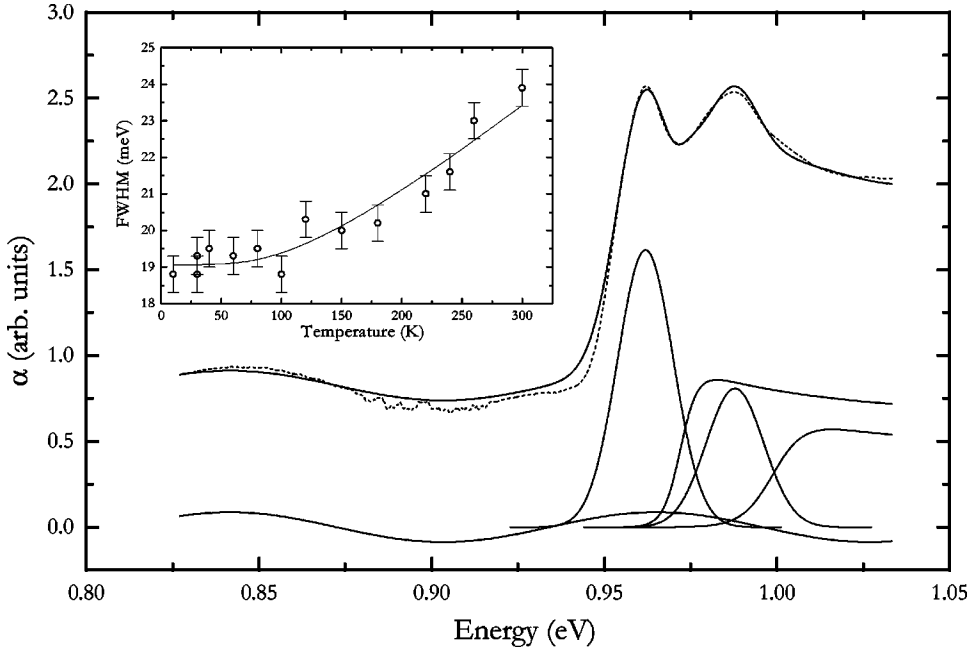

FIG. 2. Absorption spectrum at $T=10$ K for the C461 MQW showing the exciton peaks (dashed line) and the fitting result (solid line). The separated contributions to the spectrum are also reported: two Gaussian functions, two steplike functions multiplied for the Sommerfeld factor and a sinusoidal contribution, introduced to roughly take into account the artificial background modulation of the spectrum due to the interference effects. Inset: full width at half maximum (FWHM) vs temperature for the hh_1-e_1 absorption peak of the same MQW. The solid line gives the fitting curve obtained through the usual empirical law of Chemla (Ref. 29), by assuming the optical phonon energy equal to 29.8 meV. The bars indicate the uncertainty in the energy position of the peaks.

TABLE II. (Al,Ga)Sb material parameters used in the calculation of the QW transition energies. m_c m^*_{hh} m^*_{lh} are the effective masses perpendicular to the layer for electrons, heavy and light holes calculated from the Luttinger parameters. The remaining parameters were used in the evaluation of the strain effects: a is the lattice parameter, $E_{\rm av}$ is the averaged mean of the three maxima of the VB, a_v and a_c are the hydrostatic deformation potentials of the VB and CB, respectively, C_{11} and C_{12} are elastic moduli of the crystal, Δ_o is the splitting between the split-off band and the top of the VB without strain, b is the uniaxial deformation potential, $\alpha_{\rm th}$ is the coefficient of linear thermal expansion of the layer. The corresponding coefficient for GaAs was $\alpha_{{\rm th},s}=6.86\ 10^{-6}\ {\rm K}^{-1}$ (see Ref. 38).

Parameter	GaSb	AlSb	$Al_{0.4}Ga_{0.6}Sb$
E_g (eV)	0.811 [a]		1.340 [a]
m_c	0.0412	0.121	0.07312
m_{hh}	0.2197	0.329	0.2634
m_{lh}	0.0478	0.1389	0.0842
a (Å)	6.09596 [b]	6.1353 [b]	
$E_{\rm av}$	−6.25 [c]	−6.66 [c]	
a_v (eV)	0.79 [c]	1.38 [c]	
a_c (eV)	−6.85 [c]	−6.97 [c]	
Δ_o (eV)	0.82 [c]	0.65 [c]	
$C_{11}(10^{11}\ {\rm dyn/cm^2})$	8.84 [d]	8.77 [e]	
$C_{12}(10^{11}\ {\rm dyn/cm^2})$	4.03 [d]	4.34 [e]	
b (eV)	−2 [f]	−1.35 [f]	
$\alpha_{\rm th}\ (10^{-6}\ {\rm K}^{-1})$	7.75 [e]	4 [e]	

[a]References 27 and 31.
[b]Reference 23.
[c]Reference 40.
[d]Reference 41.
[e]Reference 42.
[f]Reference 38.

Figure 4 shows the experimental data as function of the *nominal* values of the well widths, after (i) and (ii) corrections: open circles refer to the C405, C461 and C462 samples; the data indicated by squares refer to the transmission spectrum of the C376 MQW, which was not taken into account in the fitting, owing to slightly different parameters (see Table I).

In the analysis the *nominal* well width dependence of $E_{hh,1}$, of $E_{lh,1}$ and of their difference $E_{lh,1}$-$E_{hh,1}$ were simultaneously fitted: the latter curve was included in the selfconsistent analysis after the ascertainment of an uneliminable discrepancy of about 1–4% between experimental and theoretical data, in order to ensure that such a discrepancy resulted as being nearly the same for all the data. The discrepancy, corresponding to an energy difference of 10–40 meV, was the same for all the MQW's and was independent either of the inclusion of the strain effects or of the variation of the band parameters in a wide range of physically reasonable values, suggesting that the effective well thicknesses are systematically lower than the nominal ones. It is worth noting that a reduction of the effective well thickness of a few monolayers has already been observed in GaSb/AlGaSb MQW's:[20,49] after an accurate SIMS and PL investigation of their samples, Lomascolo *et al.* attributed the effect to a significant Al diffusion into the well symmetrically from both the interfaces.[49] More generally, the discrepancy, together with the wide Stokes shift of the PL peak, agrees with the hypothesis of an appreciable interfacial roughness, which is not surprising in a GaSb/AlGaSb system (see, e.g., Ref. 13). According to the Dargam and Koiller calculation,[50] a 2–3 monolayers thick interface could explain the discrepancy observed in our cases. Unfortunately the x-ray characterization

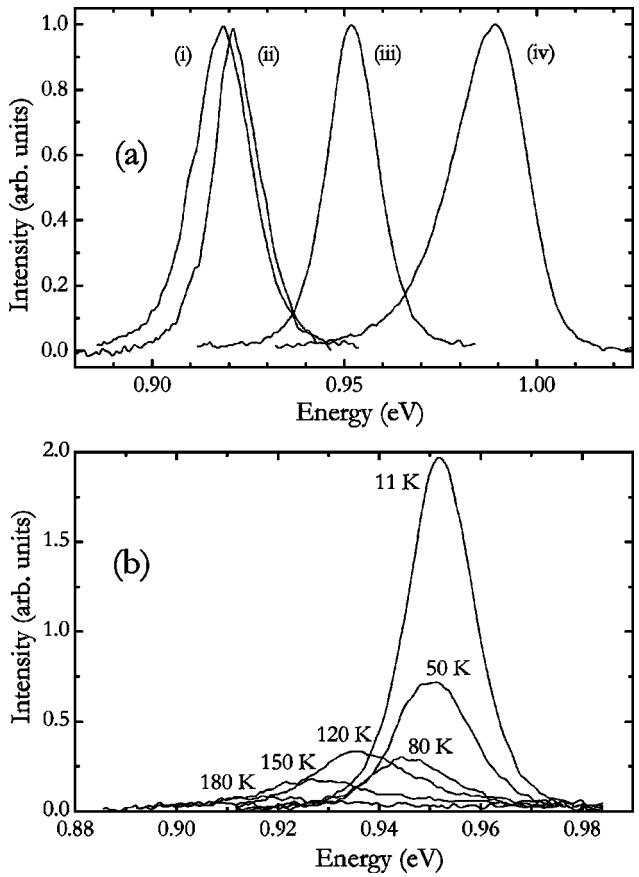

FIG. 3. (a) Photoluminescence spectra at $T=10$ K for four MQWs of different thicknesses: (i) C405 ($L_z=71$ Å), (ii) C462 ($L_z=60$ Å), (iii) C461 ($L_z=50$ Å), and (iv) C376 ($L_z=45$ Å). The peaks correspond to the intersubband hh_1-e_1 transitions. The exciting source was an argon laser ($\lambda=514.5$ nm); the power density of the incident light was about 2.5 W/cm^2 (measured on the sample). The spectral resolution of the optical system was 1.5 meV. (b) PL spectra taken at different temperatures for the C641 MQW (power density on the sample of 2.5 W/cm^2).

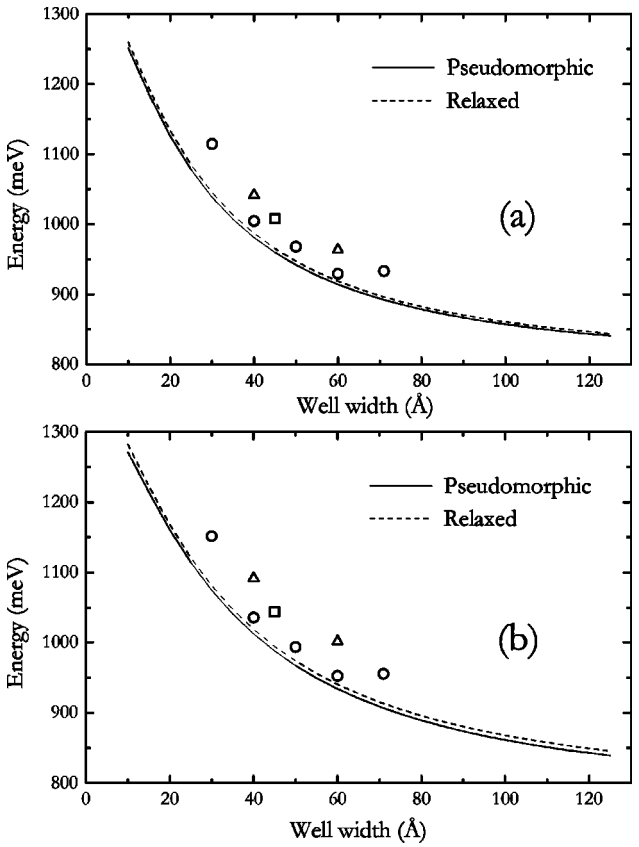

FIG. 4. $T=10$ K: energies of electronic transitions in GaSb/$Al_{0.4}Ga_{0.6}Sb$ MQWs as a function of the *nominal* well width: (a) $E_{hh,1}(L_z)$; (b) $E_{lh,1}(L_z)$. In both cases, the continuous and dashed lines refer to theoretical curves obtained under the hypothesis of pseudomorphic and relaxed structures, respectively; open circles and squares: experimental absorption data; triangles: photoreflectance data (from Ref. 20). The uncertainty of the absorption data is about ± 1 meV (not related to the dimension of the symbols).

of the structures could not confirm this hypothesis, the broadening of the rocking curve being strongly enhanced by the effects of strain relaxation at the interface between the MQW and the GaAs substrate. However, this idea is supported by a recent optical investigation performed by Ferrini *et al.*[19,20] on GaSb/AlGaSb single QW's grown on GaSb substrates through the same MBE apparatus as the present ones, at the same temperature and BEP ratio. High resolution x-ray diffraction measurements were useful in that case to show that the effective well thickness was systematically $\approx 2-3$ monolayers lower than the nominal value. The photoreflectance data relative to two QW's taken from Ref. 20 are included in Fig. 4(a) and Fig. 4(b) (triangles) vs the *nominal* well thickness for comparison with our results.

A significant interfacial roughness is also consistent with the high value of the intrinsic inhomogeneous broadening obtained from the line shape analysis of the absorption and PL peaks, $\Gamma_o^{abs}=19\pm 1$ meV and $\Gamma_o^{PL}=16.2\pm 1.5$ meV, respectively, although the value of $\Gamma_o^{PL}\approx 8$ meV reported by Ferrini *et al.*[20] for a *single* QW suggests that also the fluctuations of the well thickness in a *multiple* quantum structure and the relaxation of strain due to the use of GaAs substrate considerably contribute to the broadening of the peaks.

IV. CONCLUSIONS

An accurate analysis of the optical properties of GaSb/$Al_{0.4}Ga_{0.6}Sb$ MQWs of different well widths was performed through absorption and photoluminescence measurements at different temperatures, in the spectral range 0.7–1.3 eV. The energies of the electronic transitions between quantum states were compared with theoretical data calculated in the frame of the envelope function approximation: the best agreement was obtained for values of the band offset and nonparabolicity coefficients in good agreement with data previously reported in the literature.

The possibility to grow In-free samples of good quality without the inclusion in the structure of a buried GaSb buffer layer seems to be confirmed by the present data, encouraging the settlement of the method.

The good quality of the samples is confirmed by the well resolved details of the absorption spectra: the possibility of a reliable line shape analysis of the transmission peaks was demonstrated, supported by the fact that the experimentally determined ratio between the heavy to light ''in-plane'' re-

duced masses is in excellent agreements with the literature ones. However, the presence in all the samples of a significant interfacial roughness, not surprising in the GaSb/AlGaSb system, was invoked to explain an uneliminable discrepancy between experimental and theoretical transition energies, consistently with the wide inhomogeneous broadening of both absorption and PL peaks obtained from the line shape analysis.

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