Investigation of Roughness at InP/InAs Interfaces *

Maria J. S. P. Brasil
Instituto de Física, Universidade Estadual de Campinas
Caixa Postal 6165, Campinas, SP 13083-970, Brasil

R.. E. Nahory, Maria C. Tamargo and S. Schwarz
Bellcore, 331 Newman Springs Road, Red Bank, NJ 07701, USA

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We discuss the morphology of heteroiiiterfaces formed between phospliorus- and arsenic-based semiconductor compounds. Results of low temperature photoluminescence and Secondary Ion Mass Spectroscopy are presented as effective tools to investigate the heterointerfaces. The analysis of tlie photoluminescence results evidences roughness on different scales at the interface. The influence of tlie growth parameters, such as growth temperature and substrate orientation on the interface roughness is investigated. Details of the As-P exchange effect usually observed a.t these systems are explored with a systematic set of samples where tlie InP surface was exposecl to As following different procedures.

I. Introduction

The atomically perfect interface between two different semiconductor compounds proposecl by some models gives place, in a realistic view, to tlie rough interface real systems. Even while advances on the growtli techniques in the last decades miniinized the interface roughness to reasonable levels, it is very common that a new device or the demonstration of a new physical effect face problems generated by roughness at heterointerfaces. The detailed understanding of thc interface morphology is an endeavor not only by the desire of eliminate these problems but also by the fact that the control of the interface roughness can be a tool to build new structures[1].

There has been a growing interest in heterostructures combining InP and As alloys for various opto-electronic devices. However, previous studies suggest difficulties to obtain abrupt interfaces in these systems. These difficulties are inherent to all gas- source techniques and they are related to tlie excliange between As and P. Growth halts are usually employed to avoid having simultaneously As and P ill the growth chamber. However, it has been shown that there is a strong effect of As and P interrnixing during growth halts[2-5].

In this work we investigate the interface of thin InP/InAs quantum wells (QWs) grown by Chemical Beam Epitaxy. The advantage of this system resides on its simplicity. First it is composed of binary semiconductors and this eliminates the uncertainty of alloy fluctuations. Second, it presents a constant group III element and only the group V elements of interest, As and P, are switched at the interface. The interface morphology is investigated using photoluminescence (PL) and Secondary Ion Mass Spectroscopy (SIMS). The luminescence is characterized by multiple-line spectra and is very sensitive to the growth parameters. We discuss the interpretation of the emission lines based on long range and short range roughness as compared to the exciton diameter. The PL emission energies give an estimation of the QW thicknesses and are compared to the integrated As in the sample obtained by SIMS. The As and P exchange is investigated by exposing InP to As, which results in an effective InAs QW. We show that we can reduce the As incorporation on InP surfaces by modifying the interface chernistry and the gas

*Invited talk.

valve switch ng sequence.

II. Experimental

The samples were grown by Chemical Beam Epitaxy on singular snd vicinal (100) InP substrates tilted 2º off toward the [011] direction, using trimethylindium (TMIn), triisobutylaluminum (TIBAl), craclted arsine (As_2) and cracked phosphine (P_2). The growtli temperature was varied between 480 and 520 C. All samples consist of a 2000 Å InP buffer layer, a single InAs QW and a 200 Å InP top layer. We have used two different procedures to grow the InAs layer. In tlie first procedure the InAs was actually grown, in the sense that during the 10 sec. growth of InAs (nominal 2 monolayers) both the TMIn and the As_2 sources were open. A 2 sec. growth halt prior to and after tlie As-P switch was used to eliminate residual gases on tlie chamber. In the second procedure the InAs layer was formed by a simple exposure of the InP surface to As_2. The exposure time was 8 sec., during which both the TMIn and the P_2 sourtes were closed. The 2 sec. delay used for tlie group V switch in the first proccdure and tlie 8 sec. exposure in the secoiid one are both based on previous results witli InP/InAlAs heterojunctions[5] that have shown definite As-P exchange effects for 8 sec. exposure of InP to As, but no observable effects for times as short as 2 sec. in our growth system.

Low temperature spectra were measured at 5 K using the 488 am Ar laser line for excitation and a 0.75 m spectrometer and a cooled Ge detector, for detection. SIMS depth profile were obtained with an Atomika 3000-30 ion microprobe employing 3 keV O_2^+ bombardment at normal incidence.

III. Results and discussion

Typical low ternperature PL spectra are presented in Figs. 1 and 2. They consist of multiple lines, varying from singlets up to 8 peaks, with linewidths of the order of 20 meV. There is no detectable signal from the InP barrier layers, which shows that. tlie carriers are efficiently collected by the single InAs QWs. The vertical lines correspond to envelope function calculation for InAs QWs with integer number of monolayers, taking into account the effect of strain. Despite the expected error of tlie envelope function model for very thin QWs, this result can give an estimation of the InAs thickness formed in our samples. The comparison between the PL peaks and the calculated emission energies indicates the formation of InAs layers with thicknesses varying from 1 to 8 monolayers, in contrast to the nominal 2 monolayers.

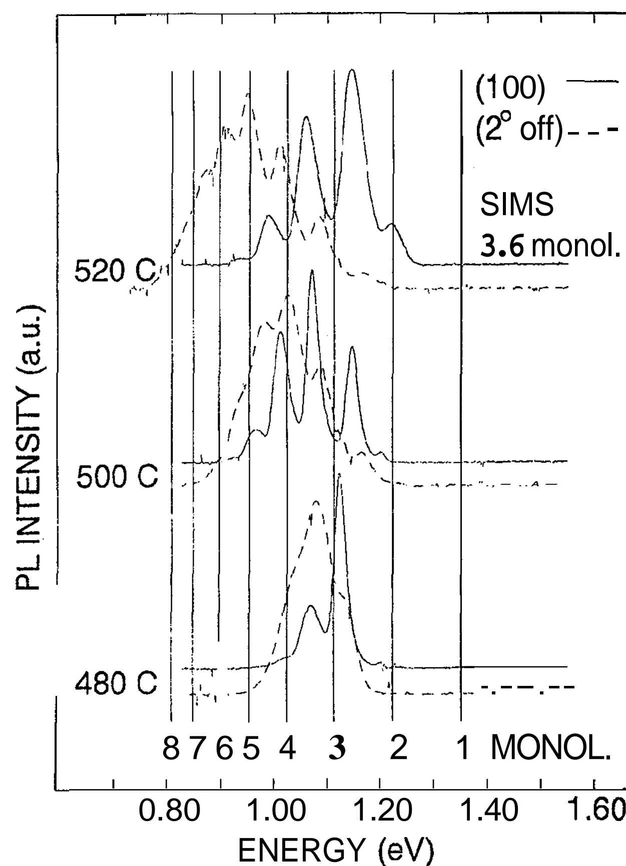

Figure 1: Low temperature PL for intentionally grown InP/InAs single QWs with a 2 monolayers nominal thickness. We used three different growth temperatures: 480, 500 and 520 C, and two different substrate orientations: (100) and 2° off (100) toward the [011] direction. Vertical lines correspond to envelope function calculation for InAs QWs with integer number of monolayers. SIMS results give the integrated number of InAs monolayers in the sample.

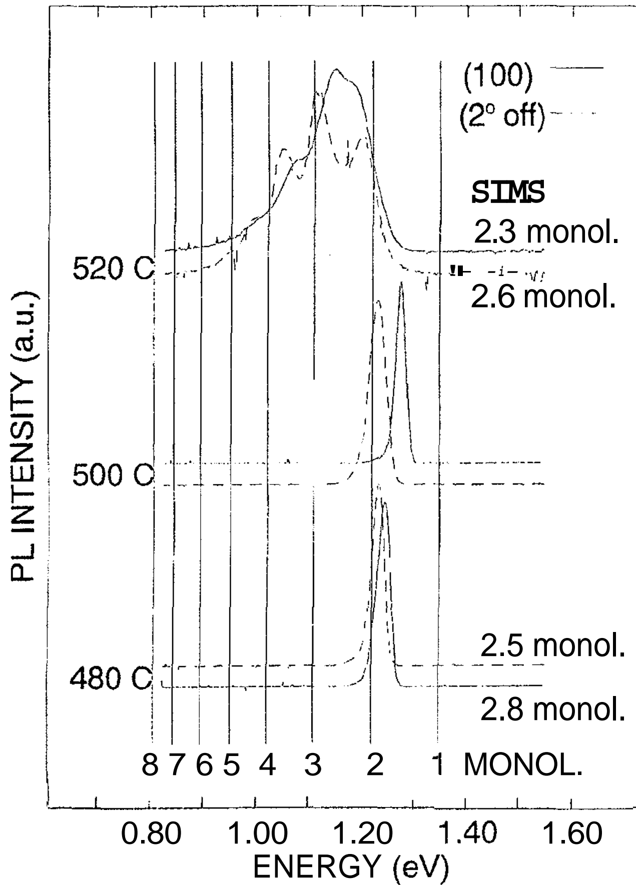

Figure 2: Low temperature PL for InP/InAs QWs obtained by exposing the InP surface to As_2 for 8 sec. We used three different growth temperatures: 480, 500 and 520 C, and two different substrate orientations: (100) and 2° off (100) toward the [011] direction. Vertical lines correspond to envelope function calculation for InAs QWs with integer number of inonolayers. SIMS results give the integrated number of InAs monolayers in the sample.

It is evident from tliese figures that tlie position of the PL peaks change significantly from sample to sample. We rcmark Iiowever that tlie variation of the peak energies as a function of the location of the laser spot for a given sample is negligible as compareci to the separation between peaks.

The rcalistic description of an interface is a. difficult task, two simple models are traditionally used. The first one is based on two perfect binaries separated by a rough edge. The roughness is classified as *microroughness* or *macroroughness*, if its length scale is short or long, respectively, as compared to the effective PL probe, tlie excitonic diameter. The scconcl model describes the inteiface as a several monolayer thick region consisting of an alloy whose composition varies from one material to the other that make up the heterointerface. The two models are basically equivalent if we consider that the alloy interfacial layer can he alternatively described by a rough edge. However, for a complete equivalence, we have to consider that both the macro and tlie microroughnesses can be extended for more than one monolayer.

To simplify our discussion, we will adopt here the model based on a rough edge. Fig. **3** shows the possible combinations of micro and macroroughnesses for a single QW and their expected PL emissions. We have considered the basic model where macroroughness is extended to various monolayers but microroughness is limitecl to only one monolayer. The presence of multiple resolved peaks instead of a single broad emission can only be explained by the presence of large InAs structures as compared to tlie exciton diameter (macroroiighness), so that different excitons probe InAs QWs with different thicknesses. The traditional interpretation of the multiple peaks is that they originate from excitonic recombination in terraces with perfect integral number of monolayers, which implies the absence of microroughness[6,7]. This interpretation has been recently contested by several authors[8-10]. In fact, since the peak energies in our spectra vary in a random way from sample to sample, it is evident that these large InAs islands do not correspond to integer numbers of monolayers. To explain the non-integer number of monolayers we have to consicler tlie existence of a microroughness superposed to a macroroughness. In this case, different excitons prohe different InAs islands with fast varying tliicknesscs due to the microroughness. The fast varying thickness can be interpreted as an effective thickness with a non-integer number of monolayers.

Recently, Carlin et al.[9] proposed a simple model to explain their experimental results. They considered that microroughness, represented hy an effective non-integer number of monolayers dm, is constant for

a given sample. In this case, due to macroroughness, the PL should give rise to multiple peaks from regions witli effective thicknesses: $1+dm$, $2+dm$, $3+dm$, ... monolayers. We point out that this model implies in different sets of emission energies for different samples with distinct values of dm, but it maintains a constant step of one-monolayer between islands in a given sample. This model was applied in Fig. 4, where tlie PL energies from various samples are plotted against tlie expected InAs thickness, maintaining the effective one-monolayer steps. For each sample, we used tlie best value of dm tliat fits the experimental results. Tlie solid line corresponds to tlie calculated emission eiiergy using the envelope function approximation. The model works reasonably well for some samples (circles), but strongly fails for others (diamonds). The failure is clue to tlie variation of tlie energy separation between two adjacent peaks, which is not the constant one-monolayer step predicted by tlie model.

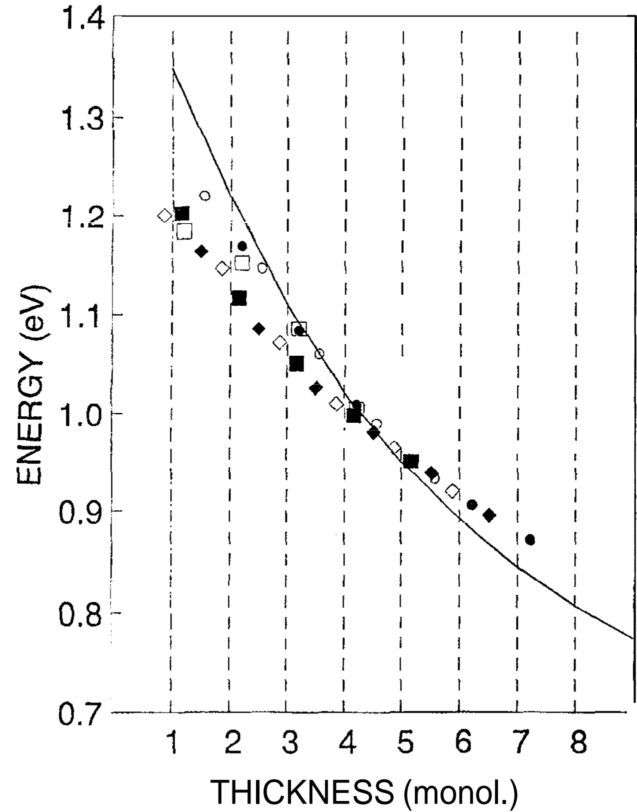

Figure 4: Energies of tlie PL peaks for various samples versus tlie QW tliickness. We considered that the InAs islands tliickness for a given sample are given by: $1+\mathrm{dm}$, $2+\mathrm{dm}$, $3+dm$, ... moiiolayers, wliere dm is the effective non-integer number of monolayers din that gives the best fits the experimental results to the tlieoretical curve: intentionally grown QWs at 520 C (circles), intentionally grown QWs at 500 C (diamonds) and QWs obtained by exposing tlie InP surface to As_2 at 520 C (squares). Open symbols correspond to (100) substrates and filled symbols, to 2° off (100) toward the [011] direction. The solid lhe corresponds to the calculated emission energy using the envelope function approximation.

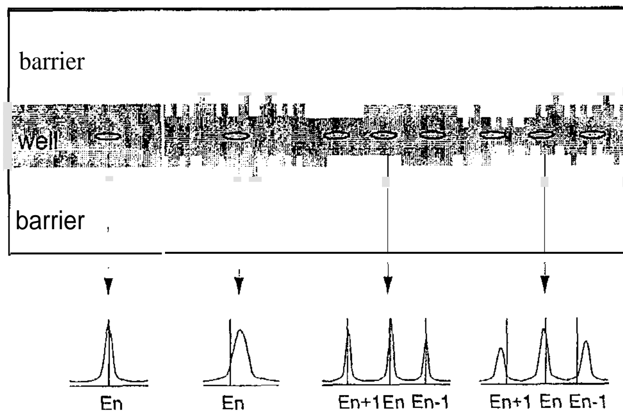

Figure 3: Sclieriatic representation of a QW with rough interfaces. We show four clifferent roughness configurations as compared to tlie excitonic diameter (represented by discs), and their expected PL emission. Tlie bars in the PL spectra correspontl to tlie energy emission for QWs with integer number of inor olayers. (a) atomically perfect interfaces, (b) microroughness, (c) macroroughness, (d) microroughness superposed to macroroughness.

Tlie conclusion of this analysis is that our InP/InAs interfaces show a superposition of micro and macroroughness, and that tlie niicroroughness is not constant over all the islands of a given sarnple. It is always possible to explain any energy emission between the QW and tlie barrier band gap using some combination of micro and macroroughnesses. Similarly, we could describe any experimental energy with an alloy interfacial layer. However, there are too many variables on the morphology of tlie InAs islands, and fitting the experimental energies for each sample using precise arrangements of micro and macro roughness, or using interfacial lay-

ers with specific variable composition, will not give any further conclusive result about the islands. We also remark that an alternative possibility to explain tlie experimental energies include an extra effect not considered in the simple envelope function approximation, such as quantum lateral confinement.

SIMS results presented in Figs. 1 and 2 give the integrated number of InAs inonolayers in the sample. Although the depth resolution of SIMS is of the order of 60 Å, which is too broad to resolve the InAs layers, we can obtain the integrated As content in our samples with estimated absolute and relative accuracies of about 20% and 10%, respectively, using InAsP calibration standards. SIMS results are reasonably close to the nominal 2 monolayers thickness. We remark that the thickness of the InAs islands estimated by tlie PL peaks indicates that some samples present islands as tliick a.s 8 inonolayers. However, in tlie special case of a PL spectrum consisting of a single peak, it should better reflect the InAs average thickness giveii by SIMS. The energy of the singlet PL spectra are indeed reasonably close to the calculated energy emission for a. 2 monolayers thick InAs QW. Briefly, SIMS results give tlie average thickness of the InAs layer while the PL spectra give information about the InAs distribution.

Figs. 1 and 2 show that the As distribution is nonuniform and strongly dependent on growth conditions. The number of PL peaks increases a.s the growth temperature is increased. This means that, a.s we increase the growth temperaturc, the InAs layer prefers to build up as thick, non-uniform islands. The growth temperature effect is observed for both procedures and substrate orientations, but it is remarkably abrupt for the exposed samples. Samples exposed to As at 520 C show up to 5 different InAs island thicknesses but all samples exposed below 520 C show only singlets emissions, suggesting a much more uniform As distribution. SIMS results show however that the total As incorporation is not very sensitive to the growth temperature. The estimated thickness for each of the exposed samples is of tlie order of 2 monolayers. A careful analysis indicates only a very small, but systematic increase of the order of 10% in the As incorporation as we vary the growth temperature from 480 to 520 C. Finally, samples grown in vicinal substrates show a slight but systematic increase in the As content as compared to the ones grown on singular substrates. This indicates that the presence of tlie step edges on vicinal substrates, where growth is expected to occur preferentially, enhances the As incorporation.

In conclusion, increasing the growth temperature has only a very small effect on the total As incorporation but strongly increases macroroughness. Previous iiivestigations about roughness at InAs/InP interfaces have shown that macroroughness also increases as the growth halt time is increased[9]. The increase of both the growtl-i temperature and the growth halt time increases the As mobility on the layer surface. This means that an increase of the As surface mobility induces InAs clustering. This behavior is probably related to the strain due to the lattice mismatch of this system which may favor the starting of a 3D growth[11,12].

To further investigate the As incorporation on the InP surface, we have performed different gas switching sequences for the As exposure procedure. Two aspects are addressed in this study. The first intends to investigate the effect of unreacted species accumulated at the InP surface on the effective incorporation of As in the sainples. The second is the effect of changing the chemistry of the surface exposed to As.

In order to analyze the first question, we introduced two growth halts on the As exposure procedure. After closing tlie TMIn valve and stopping growth, we added a 10 sec. delay during which only the phosphine valve is open. After this first delay, the phosphine valve was closed and tlie arsiiie valve was opened simultaneously and the InP exposure to As was carried out for 8 sec., as in the standard procedure. After the 8 sec. exposure, tlie arsine valve was closed and the phosphine valve was opened simultaneously. We then introduced a second

10 sec. delay during which only the phosphine valve was openecl, after which, the TMIn valve mas opened resuming InP growth. The first delay is intended to minimize the presence of an excess In species on the sample surface prior to the As exposure, and the second, to minimize the an excess of unreacted As species on the sample surface after As exposure. Both species may incorporate on the subsequent growth steps, producing an tiffective amount of InAs. The PL emission from the sample following this special As exposure is presented in Fig. 5(b). This procedure was performed at 520 C, the same temperature as tlie standard As exposure sample presented in Fig. 5(a). We observe an energy shift of the PL peaks to higher energies, consistent with a decrease of the average InAs thickness. SIMS measurements support this result, giving 0.9 ML to the InAs layer grown on singular substrate and 1.8 ML for tlie vicinal one. Since we minimized the excess unreacted species on the sample surface, tliis value should reflect the effective As-P exchange effect. We remark tliat the number of PL peaks decreases with the introduction of the growth delays, inclicating that they are also effective to eliminate the formation of macroroughness. Finally, the effect of enhanced As incorporation on vicinal substrate as compared to singular ones is still observed, indicating that the presence of step edges assists the As-P exchange.

To investigate the chemistry surface effect we have grown samples where an AlP layer, nominal one-monolayer thick, was added prior to the As exposure. PL spectra from these samples are shown in Figs. 5(c) and 5(d). The choice of an Al-rich surface is based on previous results that have shown an effective stabilization of P- compound interfaces using this element[5]. This can be understood on the basis of a stronger AlP-chemical bound as compared to the InP, wliich is reflected on a higher congruent evaporation temperature of AlP (> 700 C) as compared to the InP (360 C)[13]. The sample presented in Fig. 5(c) corresponds to the standard 8 sec. As exposure of the AlP surface at 520 C, with no growth halt delays. We have not performed SIMS measurements for this sample, but its PL emission shows a energy shift to higher energies as compared to the standard InP exposure sample (Fig. 5(a)), indicating a decrease of the total As incorporation, as expected. The presence of the stronger AlP bounds at thc exposed surface also seems to be very effective on reducing the InAs clustering, since the PL emission from this sample is a singlet in contrast to the multiple emission peaks obtained for the InP exposure at similar conditions.

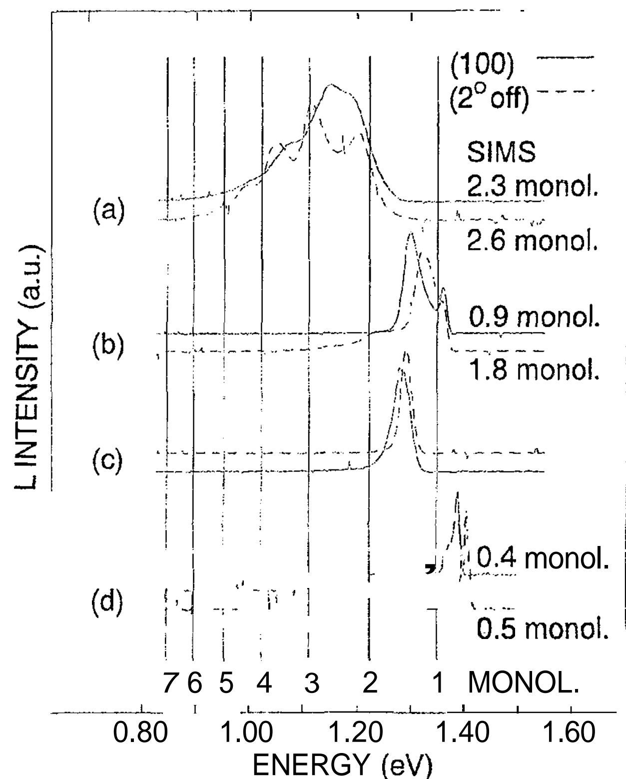

Figure 5: Low temperature PL for InP/InAs QWs obtained by exposure to As2 for 8 sec at 520 C, using two different substrate orientations: (100) and 2° off (100) toward the [011] direction. (a) InP exposure with no growtli halts. (b) InP exposure witli 10 sec. growtli halts with P, prior to aud after, the As exposure. (c) AlP exposure with no growtli halts (b) AlP exposure with 10 sec. growtli halts witli P, prior to and after, the As exposure. Vertical lines correspond to envelope function calculation for InAs QWs witli integer number of monolayers. SIMS results give the integrated number of InAs monolayers in tbe sample.

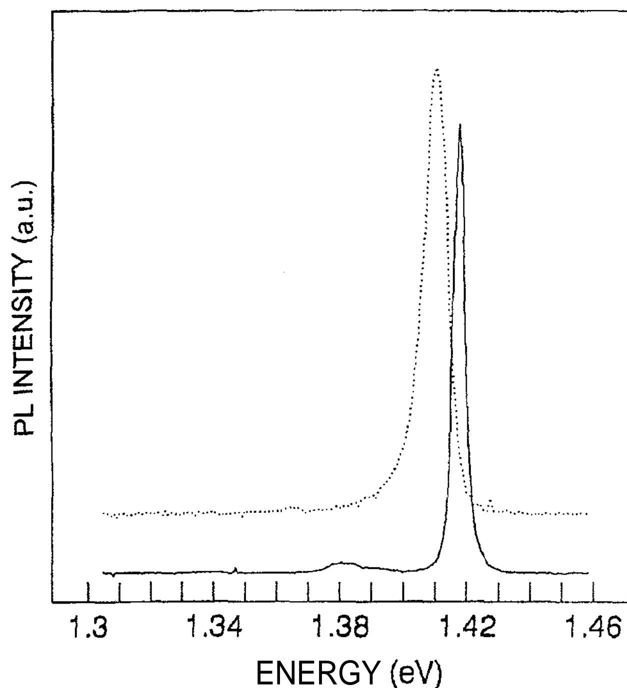

Figure 6: Low temperatuse PL for an InP/InAs QW obtained by exposing AlP to As for 8 sec., using 10 sec. growth halts witli P, prior to and after, the As exposure (dotted line), and for a reference sample consisting only of an InP buffer layer (solid line).

Finally, Fig.5(d) shows tlie PL emission from AlP exposures to As using 10 sec. growth halts with P, prior to and after, the As exposure. As discussed before, tlie growth delays are expcctecl to minimize the effcct of InAs formation clue to unreacted species on tlie surface, so that, in tliis case, we can directly compare the As-P exchange on AlP (Fig. 5(d)) and InP (Fig.5(b)). The observed PL emission shift shows that the Al-rich layer reduces tlie As-P excliange. SIRIS measurements give an integrated 0.5 ML of InAs, which supports that very little InAs is formed by As-P exchange on the AlP surface. Tliis non-zero value is however strong evidence tliat some As is still present on these samples. The PL spectra for tliese special samples are markedly different from our typical InAs QWs. The energy emission is very close to the InP bandgap energy and the linewidth is relatively small (of the order of 10 meV). Fig. 6 shows tlie comparison of the PL from one of these sample with tlie PL spectra of a sample consisting only of an InP buffer layer. It is remarkable that this less-than-one-monolayer InAs layer is so efficient on collecting all the pliotogenerated carriers, since there is still no measurable signal at the InP bandedge.

IV. Conclusions

The PL emission from thin QWs is a useful tool to qualitatively investigate semiconductor heterointerfaces. Our PL measurements on thin InAs/InP QWs presented multiple peaks corresponding to InAs islands with non-iiiteger number of monolayers. We interpret tliis result as evidence tliat tlie InP/InAs interfaces present a. superposition of micro and macroroughness and tliat microroughness is not constant over all tlie islands of a given sample.

SIMS results give the integrated content of As, or tlie averaged number of InAs monolayers, on the sample. Tliis value agrees within the experimental error with the nominal value of the InAs thickness as deduced from growth conditions. PL spectra on the other hand reveal the distribution of incorporated As. We observed tliat increasing the growth temperature sliglitly decreases tlie As incorporation, but strongly increases the macroroughness. Resides, tlie presence of step edges on vicinal substrates slightly enhances As incorporation.

A consistent set of samples where the InP surface was exposcd to As showed tliat InAs layers are fornied by two effects: capture and posterior incorporation of unreacted species, and actual As-P exchange. We have also shown tliat an Al-rich surface maltes tlie P-compound interface much more stable, reducing both the As incorporation and tlie InAs clustering.

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