Photoluminescence study of undoped and modulation-doped pseudomorphic $Al_yGa_{1-y}As/In_xGa_{1-x}As/Al_yGa_{1-y}As$ single quantum wells

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Photoluminescence has been used to describe the dominant recombination processes in undoped pseudomorphic $GaAs/In_xGa_{1-x}As/GaAs$ single quantum wells for $x<0.17$. The temperature and intensity dependences of the photoluminescence indicate emission arising from both donor-bound and free excitons in all well widths studied (6–160 Å). No evidence for the presence of monolayer fluctuations has been seen despite the high optical quality of the excitonic features. By recording photoluminescence excitation spectra as a function of well width it has been possible to unequivocally identify peaked excitonic absorption spectra with absorption transitions involving light-hole states. Comparisons of absorption energies with calculations of confined levels that include strain suggest that the light-hole state is not confined in the $In_xGa_{1-x}As$ well. Modulation-doped $Al_yGa_{1-y}As/In_xGa_{1-x}As/GaAs$ structures are shown to be ideal systems for studying the optical properties in the presence of free carriers. With increasing carrier concentration the excitonic luminescence from the well broadens, and at the highest carrier concentrations occupation of $n>1$ conduction-band states is clearly revealed by the presence of a high-energy emission.

I. INTRODUCTION

It has recently been shown that $GaAs/In_xGa_{1-x}As/GaAs$ single quantum wells (SQW's) have optical qualities comparable to those of lattice-matched $Al_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ SQW's,[1] despite the strain present in the QW due to the different lattice constants of the two components. Low-temperature photoluminescence (PL) has given information on the interface quality in SQW's. In an $AL_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ SQW, the linewidth of the PL peak associated with the 1*e*-1hh excitonic emission (hh denotes heavy hole, lh light hole) is a measure of the heterojunction interfacial quality.[2] Reported here for $GaAs/In_{0.11}Ga_{0.89}As/GaAs$ SQW's with well widths below 40 Å are full widths at half maximum (FWHM) of the PL that decrease with decreasing well width, with a very small value of 0.23 meV observed for a 6-Å well. The PL linewidth-broadening mechanisms in the $GaAs/In_xGa_{1-x}As/GaAs$ system are likely to be different from those in the $Al_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ system due to the different position of the ternary material with respect to the barriers and well. Indeed, for the $GaAs/In_xGa_{1-x}As/GaAs$ strained-layer system it seems that the FWHM of the excitonic PL is almost independent of the well width below the critical thickness for tetragonal distortion, in sharp contrast to the $Al_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ system.[2]

Photoluminescence-excitation (PLE) spectra provide detailed information about excitonic interactions with heterojunction interfaces, as well as band-structure effects,[3] and PLE results have been used to obtain values for the conduction and valence-band offsets. The one analyzed published PLE result from $GaAs/In_{0.05}Ga_{0.95}As/GaAs$ multiple quantum wells (MQW's) suggested that most of the band offset in this system occurred in the valence band,[4] but other workers have reached the opposite conclusion using PL measurements as a function of quantum-well widths.[5] In this paper it is shown that confined-state energies can be obtained from photoconductivity spectra (PCS), using a simple coplanar electrode geometry, which complement PLE measurements as a function of well width.

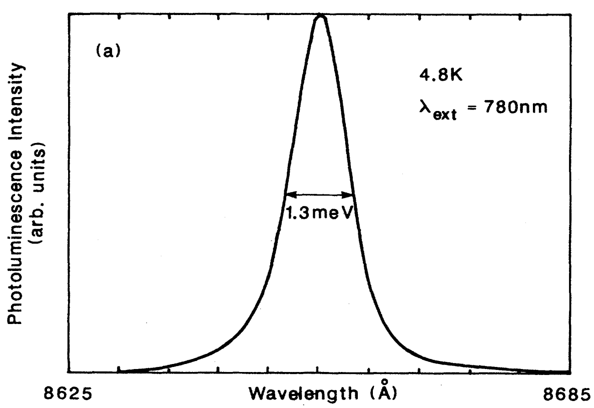
Details of molecular-beam-epitaxy (MBE) growth and experimental procedures are outlined in Sec. II. Section III is concerned with a discussion of PL and PLE measurements on undoped $Al_yGa_{1-y}As/In_xGa_{1-x}As/Al_yGa_{1-y}As$ SQW's. Section IV discusses PL results on modulation-doped *n*-type $Al_yGa_{1-y}As/In_xGa_{1-x}As/GaAs$. It is shown that this system is almost ideal for studying the relative importance of excitonic and free-carrier effects.

II. EXPERIMENT

The samples used in this study were grown by molecular-beam epitaxy on semi-insulating GaAs (100) substrates. Most of the undoped samples comprised a 0.5-μm GaAs buffer, an $In_{0.11}Ga_{0.89}As$ well, and a 0.5-μm GaAs cap; the exception was the 150-Å SQW sample, with a 1-μm buffer layer. The growth temperatures for the GaAs and $In_xGa_{1-x}As$ layers were 580 and 540 °C, respectively, as measured using an Ircon V series infrared pyrometer. To ensure that the growth temperature of the $In_xGa_{1-x}As$ was stable at 540 °C, the substrate temperature was ramped down from 580 °C over the last 350 Å of the GaAs buffer layer at 0.3 °C/s. $Al_yGa_{1-y}As/In_xGa_{1-x}As/GaAs$ and $Al_yGa_{1-y}As/In_xGa_{1-x}As/Al_yGa_{1-y}As$ SQW's were also grown. In this case the buffer and cap were grown at 620 °C. $In_xGa_{1-x}As$ growth conditions were calibrated by measuring the composition using x rays and Raman scattering.[6] PL measurements were performed using a Spex 1404 monochromator. An argon-ion laser or tungsten lamp and/or minimate combination was used for exciting the PL with the latter also used for the PCS and PLE measurements. The indium contacts used for photoconductivity measurements were soldered and then alloyed in a hydrogen atmosphere at 390 °C for 10 min before being tested for Ohmicity.

III. UNDOPED $GaAs/In_xGa_{1-x}As/GaAs$ SQW

An example of a typical PL spectrum observed from a 100-Å $GaAs/In_{0.11}Ga_{0.89}As/GaAs$ SQW in this study is shown in Fig. 1. Although under the excitation conditions used the exciting light is absorbed in the GaAs capping layer, the quantum-well PL is over 2 orders of magnitude larger than that from the GaAs capping layer, implying that the quantum wells are extremely efficient recombination centers. The quantum-well PL in Fig. 1 is very narrow (~1 meV), which is typical of this material system for wells greater than 50 Å.[1,7] Figure 1(b) displays the photoluminescence on a logarithmic versus linear scale, which brings out the common feature of the quantum-well PL in this work; that is, it is double peaked. This was observed for all well widths studied (6–160 Å). Under the low-intensity excitation conditions used to record the data in Fig. 1 the high-energy peak can only be seen clearly on a logarithmic scale; however, under appropriate temperature and intensity conditions the two peaks are clearly visible on a linear scale. The intensity dependence of such structures is displayed in Fig. 2. The higher-energy peak grows in intensity relative to the lower-energy peak with increasing power intensity. With decreasing well width the two peaks evi-



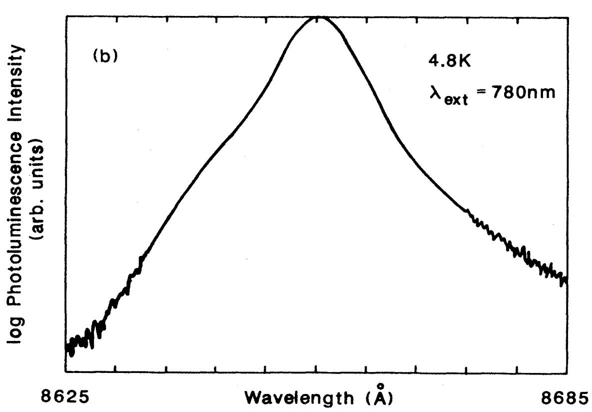


FIG. 1. Photoluminescence from 100-Å $GaAs/In_{0.11}Ga_{0.89}As/GaAs$ SQW: (a) linear intensity scale; (b) logarithmic intensity scale.

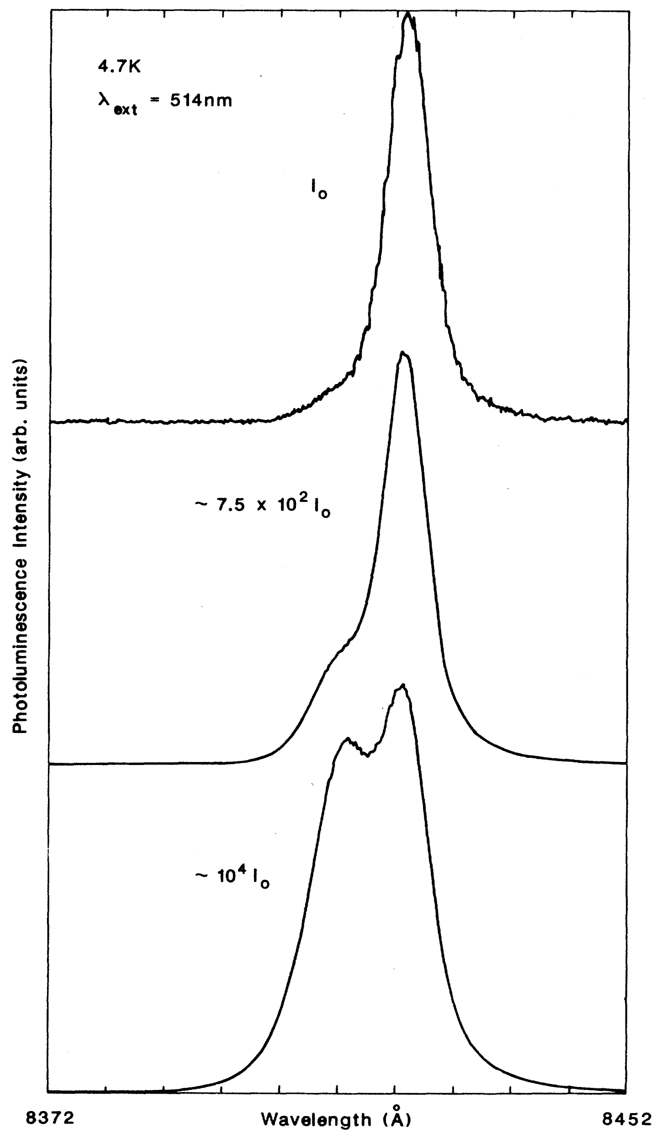


FIG. 2. Photoluminescence spectra from 60-Å $GaAs/In_{0.11}Ga_{0.89}As/GaAs$ SQW at different excitation intensities. With increasing intensity the high-energy peak grows with respect to the lower-energy peak.

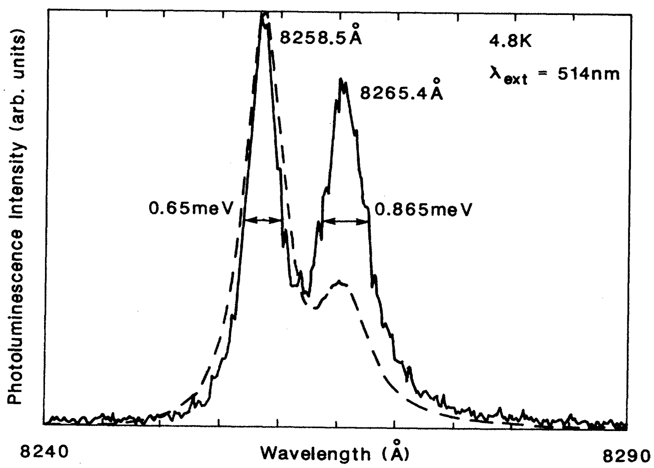


FIG. 3. Photoluminescence spectra from 20-Å GaAs/$In_{0.11}Ga_{0.89}As$/GaAs SQW at high (dotted line) and low excitation (solid line) intensities.

dent in Fig. 2 become better resolved due primarily to the decrease in linewidth, as is shown in Fig. 3 for a 20-Å well.

There are several possible explanations for the observed structure. In $Al_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ SQW it is known that interlayer fluctuations can give rise to emissions at different energies, the differences in well thickness causing the energies of the confined levels to vary.[8] However, calculations described later reveal that the energy separation between 1*e*-1hh levels caused by a well-width fluctuation of one monolayer in a 20-Å GaAs/$In_{0.11}Ga_{0.89}As$/GaAs well is ~4.2 meV. This is significantly larger than the ~1.2 meV separation between the PL peaks in Fig. 3, and the separation does not vary greatly with well width, as would be expected from monolayer fluctuations.

Another explanation is that the high-energy peak is due to free-exciton emission and the low-energy peak results from biexciton recombination, this having being assigned to almost identical PL structure from $Al_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ SQW's.[9] However, the intensity dependence of the PL emission in Fig. 2 does not correspond to that of biexcitons, and would seem to rule out this explanation.

The most likely explanation of the two-peaked structure is that the high-energy peak results from free-exciton emission, while, following the arguments of Normua, *et al.*[10] the low-energy peak is probably due to an exciton bound to a neutral donor. The relative intensity dependence of the two-peaked structure is the same as that observed from free-exciton and donor-bound-exciton emission in bulk GaAs. For the low indium concentrations used in this study, the additional binding energy due to confinement is very small, ≤ 1 meV,[11] consistent with our findings that the separation between the free-exciton and donor-bound PL peaks is a weak function of well thickness. Below 40 Å the linewidth of both the free-exciton and donor-related emission decreases.

In the absence of a dominating interfacial scattering mechanism, it is likely that the factors contributing to the linewidth in the GaAs/$In_xGa_{1-x}As$/GaAs SQW system are very different from those in $Al_yGa_{1-y}As$/GaAs/$Al_yGa_{1-y}As$. In the latter, the PL linewidth increases rapidly as the well width decreases below 80 Å.[2] For the system under study here, no increase in the linewidth is seen with decreasing well width (Figs. 3 and 4). In fact, the linewidth decreases below 40 Å. For well

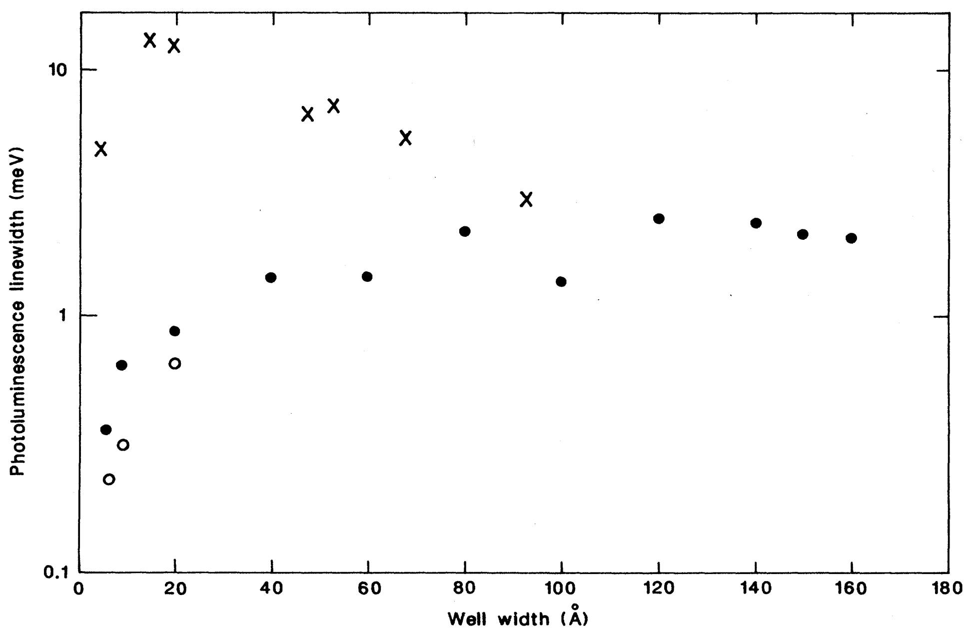


FIG. 4. Comparison of quantum-well photoluminescence linewidth (FWHM) from $Al_{0.38}Ga_{0.62}As/GaAs/Al_{0.38}Ga_{0.62}As$ wells (×) and GaAs/$In_{0.11}Ga_{0.89}As$/GaAs SQW's (○,● represent the FWHM of the free-exciton and donor-bound-exciton emission, respectively). Data for $Al_{0.38}Ga_{0.62}As/GaAs/Al_{0.38}Ga_{0.62}As$ wells from Bertolet *et al.* (Ref. 20).

widths of 40 Å and above, the data in Fig. 4 are based on the linewidth of the donor-bound-exciton emission since this dominates at the low intensities used for the measurements. In order to be able to measure the linewidth of the free-exciton emission for well widths above 40 Å, it was necessary to develop appropriate deconvolution techniques since the two peaks were often well merged (see Fig. 1). It was found that good fits to the PL spectra could be obtained when a sum function of Gaussian and Lorentzians was used for each peak. In general, the high-energy peak, arising from free-exciton recombination, was weighted towards a Gaussian, whereas the low-energy peak was Lorentzian. Using this deconvolution method it was found that above 40 Å the linewidth of the free-exciton emission was also independent of well thickness. The essential difference between the $Al_yGa_{1-y}As$/GaAs/$Al_yGa_{1-y}As$ and GaAs/$In_xGa_{1-x}As$/GaAs systems is the composition of the barriers and wells, the position of the ternary material relative to the quantum well being exchanged. The effect of this difference between the two cases should be evident towards the extremes of well thickness, where the wave function of the quantum-well exciton is confined either in the well for large well widths, or in the barrier for short well widths. The decreasing linewidth of the free-exciton emission below 40 Å in the GaAs/$In_xGa_{1-x}As$/GaAs SQW is due to a decreasing contribution from alloy scattering as the exciton wave function penetrates into the ordered GaAs binary barrier. Further information on the factors contributing to the linewidth is provided by PL spectra measurements on symmetrical $Al_yGa_{1-y}As$/$In_xGa_{1-x}As$/$Al_yGa_{1-y}As$ and inverted GaAs/$In_xGa_{1-x}As$/$Al_yGa_{1-y}As$ structures. Typical spectra are displayed in Fig. 5. The linewidth of the PL spectra from the inverted and symmetrical $Al_yGa_{1-y}As$ structures are significantly greater than when only GaAs forms the barriers. They are also greater than that obtained from $Al_yGa_{1-y}As$/$In_xGa_{1-x}As$/GaAs SQW's of comparable well thickness.[7] This indicates that additional alloy scattering occurs when ternary $In_xGa_{1-x}As$ is grown on ternary $Al_yGa_{1-y}As$. It is surprising that monolayer fluctuations are not identified by PL in this system, as narrow PL linewidths mean that they should, if present, be visible. This suggests perhaps that the presence of strain in this pseudomorphic system does not allow terracing of the quantum-well widths, since an $In_xGa_{1-x}As$ unit cell would have to be constrained along two axes; Poisson's extension allows only one axis to be constrained.

In order to obtain further information on confined states, we measured PLE spectra on a range of SQW widths. Extensive measurements of PLE spectra in the $Al_yGa_{1-y}As$/GaAs/$Al_yGa_{1-y}As$ system has led to information on band-offset energies and exciton binding energies.[3] The previous interpretation of absorption[12] and PLE (Ref. 4) measurements on GaAs/$In_xGa_{1-x}As$/GaAs SQW's and MQW's has led to different conclusions about the distributions of the band-gap differences between the valence and conduction bands and also about the nature of the light-hole transition. The first PLE measurement by Marzin *et al.*[13] displayed a steplike ab-

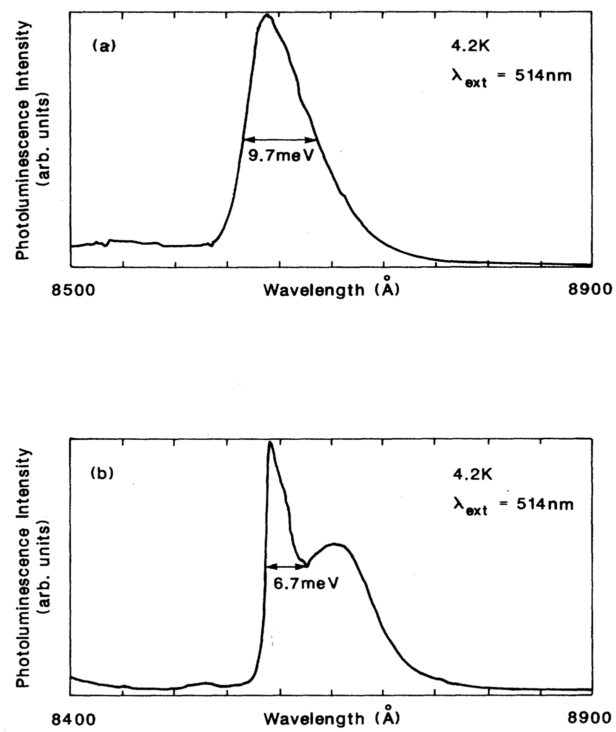


FIG. 5. Photoluminescence spectra observed in (a) 150-Å $Al_{0.15}Ga_{0.85}As/In_{0.11}Ga_{0.89}As/Al_{0.15}Ga_{0.85}As$ SQW and (b) inverted 150-Å $GaAs/In_{0.11}Ga_{0.89}As/Al_{0.23}Ga_{0.77}As$ SQW structure.

sorption for the 1*e*-1lh transition that was expected for a light hole not confined in the $In_xGa_{1-x}As$ well. This was necessarily consistent with a band offset $Q > 0.5$, where Q is the conduction-band-offset value divided by the difference in band gaps of the materials making up the heterojunction. (A similar conclusion was made and the same steplike absorption feature observed by Ji *et al.*[12] using transmission measurements.) A steplike absorption was expected for a transition between a continuum of states and a discrete state. However, recent PLE and Raman results by Menendez *et al.*[4] on a MQW structure consisting of $GaAs/In_{0.05}Ga_{0.95}As/GaAs$ suggested a band-offset ratio Q of ~ 0.4, with the light hole being confined in the $In_{0.05}Ga_{0.95}As$ wells. In this case an absorption peak was assigned to the 1*e*-1lh transition.

In addition to PLE measurements, we have used photoconductivity spectral measurements to obtain information on confined states. That the PLE and PCS techniques both measure absorption transitions between confined levels is evident from Fig. 6, where the two responses from the 100-Å identical quantum-well sample show similar structure. The PCS technique has the advantage of a greater signal-to-noise ratio and the ability to easily measure the 1*e*-1hh absorption transition. Previous photoconductivity measurements have been performed using Schottky-barrier structures, whereas the results presented here were taken using coplanar Ohmic

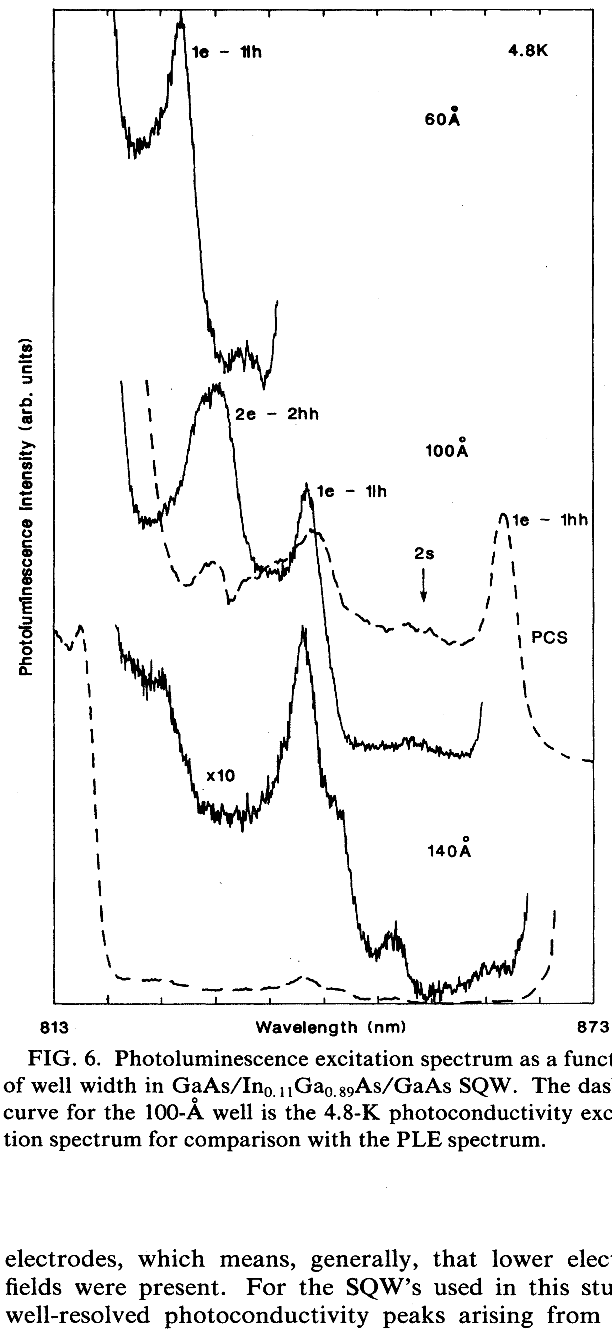


FIG. 6. Photoluminescence excitation spectrum as a function of well width in GaAs/$In_{0.11}Ga_{0.89}As$/GaAs SQW. The dashed curve for the 100-Å well is the 4.8-K photoconductivity excitation spectrum for comparison with the PLE spectrum.

electrodes, which means, generally, that lower electric fields were present. For the SQW's used in this study, well-resolved photoconductivity peaks arising from absorption between confined levels were only observed below 200 K. Often at 77 K the transitions were obscured by the presence of a persistent photocurrent in the semi-insulating GaAs substrate. The fact that a photocurrent signal arising from absorption into confined levels is observed at 4 K suggests that free excitons and free carriers coexist in the wells.

The PCS and PLE results presented in this paper were interpreted in terms of a model used by Ji *et al.*[12] for calculating confined-energy states in strained-layer GaAs/$In_xGa_{1-x}As$/GaAs quantum wells. In this model the strain-dependent band gap is calculated and incorporated into confined-energy-levels calculations. When growing on a GaAs buffer, the $In_xGa_{1-x}As$ layers are under biaxial in-plane compression, and there is a corresponding extension along the ⟨001⟩ growth direction.

The parameters of GaAs and InAs used here are the values used by Ji *et al.*[12] After obtaining the band gap for heavy and light holes using the equations described in Ji *et al.*,[12] the confined energies are calculated using a transfer-scattering-matrix technique which matches the gradient of the wave function at each heterojunction interface. In this work routines have been developed that optimize the fitting of observed PLE transitions by varying the quantum-well thickness Q and the composition, x. L was varied 5% around the value measured by transmission electron microscopy (TEM). A tendency was observed during the running of these fitting algorithms that the use of lower compositions was necessary for thin wells, despite the fact that nominally the same composition was aimed at during MBE growth.

Figure 6 shows the recorded PLE spectra from GaAs/$In_{0.11}Ga_{0.89}As$/GaAs SQW's having well widths of 60 Å, 100 Å, and 140 Å. The development of the absorption structures with increasing well width is consistent with the expected change in the number of confined levels with increasing well width. In general, with the low-resolution excitation source used in this study for the PLE measurements it was not possible to measure the $1e$-1hh absorption. For samples in which this was possible, a small Stokes shift, ≤ 1 meV, was found for $x=0.11$. The Stokes shift was found to increase for larger indium concentrations, which is typical of the trend observed with increasing Al mole fraction in the $Al_yGA_{1-y}As/GaAs/Al_yGa_{1-y}As$ system.[14] PLE measurements as a function of well width have the advantage that the confined states can be clearly identified. For instance, for wells below 100 Å for $x \leq 0.11$ only one confined conduction-band state exists, and the expected absorption transitions are $1e$-1hh and $1e$-1lh. The $1e$-1lh is clearly identified at 827.5 nm in Fig. 6 for the 60-Å well. The line shape of the $1e$-1lh absorption in Fig. 6 is similar to that from absorption processes definitely involving two confined states (e.g., $2e$-2hh, Fig. 6). The form of the $1e$-1lh absorption seen in PLE does not noticeably change with well width or composition. This absorption shape for the $1e$-1lh transition cannot be taken as simple evidence that the $1e$-1lh exciton is confined in the $In_xGa_{1-x}As$ well. The calculations described above, for all samples measured here, give a band-offset value that would lead to the light-hole state being unconfined (i.e., $Q > 0.5$). The "best fit" to the data generally gives $Q \sim 0.6$. The picture for the observed $1e$-1lh exciton absorption that we end up with is of an exciton bound at the heterojunction interface, possibly mediated by the same donor observed in the steady-state photoluminescence spectrum. The situation must be different than that observed in a type-II superlattice, such as AlAs/GaAs, where the absorption involving $n=1$ electrons at the X minimum of the AlAs and the $n=1$ heavy holes in the GaAs is greatly reduced in strength compared to type-I structures, due to the carriers being separated.[15] The increased absorption transition probability in the present case might be aided by the greater oscillator strength of excitonic absorption involving defects.

IV. PL AND PLE RESULTS ON DOPED SQW's

The effects of the presence of background carriers on the photoluminescence properties of quantum-well structures can be studied by modulation doping or under intense optical excitation. In the first case there is an excess concentration of electrons or holes, while in the second both excess electrons and holes are both present. In the case of n-type modulation-doped samples, under the situation where the concentration of photoproduced holes is small, emission is expected to involve the $n=1$ heavy-hole confined state. A full description of the optical properties of a modulation-doped system has yet to appear, with the relative importance of excitonic and free-carrier effects being unclear. In this section PL and PLE results on n-type $Al_yGa_{1-y}As/In_xGa_{1-x}As/GaAs$ are presented for a range of sheet carrier concentrations.

Figure 7 shows the PL spectra recorded at 4 K for two samples with relatively low sheet carrier concentrations. The sample structures are described in the figure caption. The PL linewidths are considerably increased upon doping, in line with previous reports.[16–18] The overall shapes of the two PL spectra in Fig. 7 are very similar: a fast rise of PL intensity from low energy followed by a slow falloff, and then a faster decrease. The high-energy, fast falloff of the intensity decreases exponentially with energy, as expected for a PL process whose spectrum is determined by a Boltzmann distribution of carriers. The energy position of the start of the high-energy, fast falloff varies with the carrier concentration, and gives an accurate measure of the Fermi level in this system. The form of the spectra in Fig. 7 is almost identical to that described theoretically by Lyo and Jones.[19] Their description was based on the PL linewidth in the doped system being determined primarily from ionized-impurity scattering of the majority and minority carriers, with smaller contributions from the thermal distribution of the carriers. In this model, excitonic recombination is considered unimportant due to screening effects. Although the shapes of the PL spectra in Fig. 7 are consistent with the dominant recombination being due to free carriers, the PLE spectrum in Fig. 8 reveals that remnants of excitonic absorption occur up to relatively high carrier concentrations. In the $Al_yGa_{1-y}As/GaAs/Al_yGa_{1-y}As$ quantum-well system it has been suggested that in a modulation-doped system the energy spacing between the absorption threshold and 1e-1hh can be described by:[16–18]

$$E_F(1+m_e^*/m_h^*)\ , \tag{1}$$

where E_F is the Fermi energy of the two-dimensional (2D) plasma, and m_e^*, and m_h^* are the effective masses for the conduction and valence bands, respectively. This equation represents the separation between the energy gap 1e-1hh and the onset of absorption in optical processes associated with a degenerate semiconductor electron plasma. The absorption edge for the excitation spectrum of Fig. 8 can be determined as the wavelength at which the intensity reaches half of its maximum value (at 845 nm in Fig. 8). It is found, however, that the Fermi level determined by substitution in Eq. 1 is different from that obtained from the photoluminescence spectrum. These differences are not understood at present.

With increased sheet carrier concentrations, the PL spectra change considerably. A new, sharp PL peak occurs at high energy [Fig. 9(a)], while the low-energy peak subsides. At the highest carrier concentrations, only a broadened high-energy peak is observed. The PLE

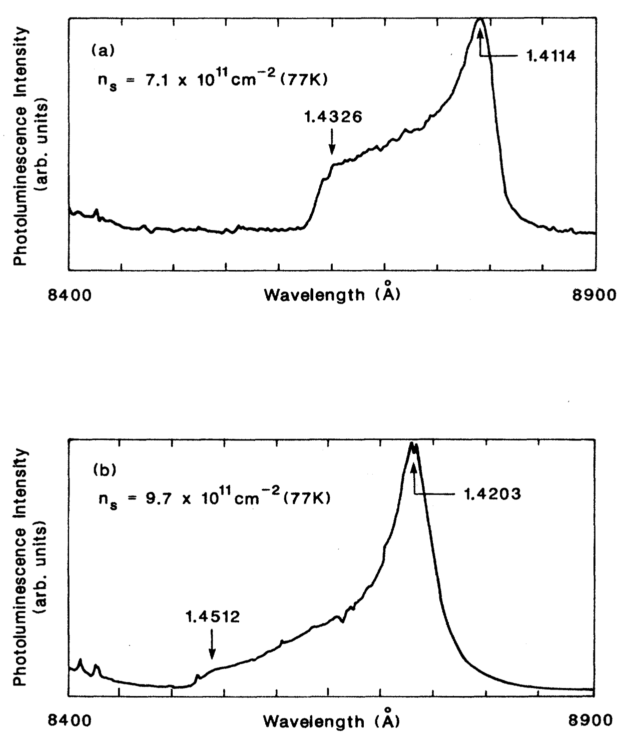


FIG. 7. Photoluminescence spectra observed from two n-type modulation-doped 150-Å $Al_{0.23}Ga_{0.77}As/In_{0.11}Ga_{0.89}As/GaAs$ SQW's having differing sheet carrier concentration. (a) $n_s=7.1\times10^{11}\text{cm}^{-2}$ (77 K). (b) $n_s=9.7\times10^{11}\ \text{cm}^{-2}$ (77 K).

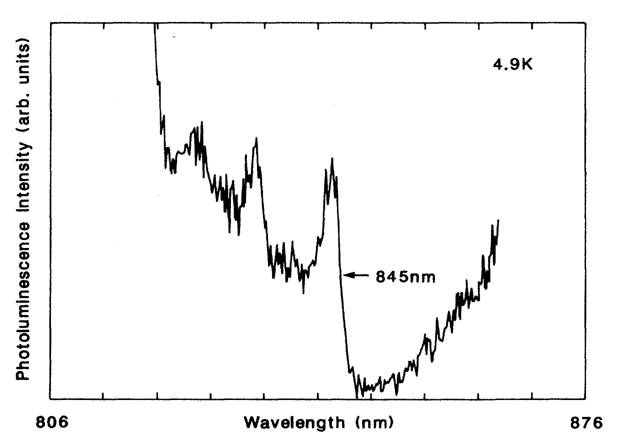
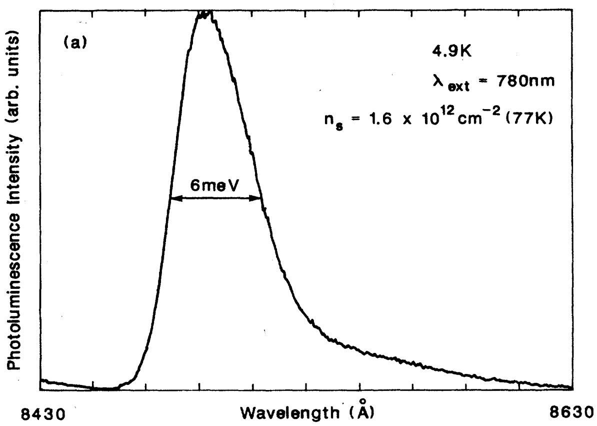


FIG. 8. PLE spectrum observed by monitoring the PL of Fig. 7(b) at 8725 Å.



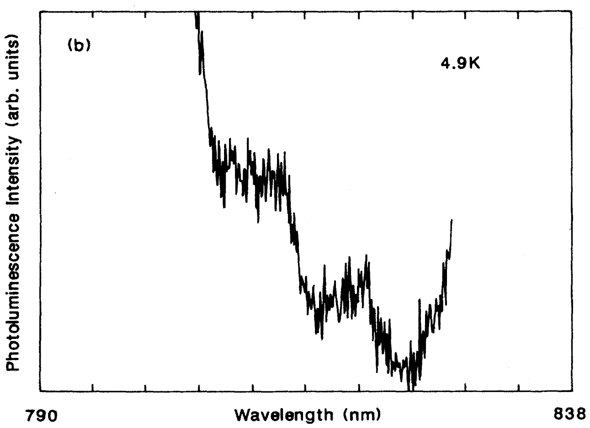


FIG. 9. (a) PL spectrum observed in n-type modulation-doped sample having a sheet carrier concentration of 1.6×10^{12} cm^{-2} at 77 K. (b) PLE spectrum recorded by monitoring the PL of (a) at 8490 Å.

spectrum for this sample [Fig. 9(b)] shows a step-like absorption with little excitonic absorption present. It is likely that the high-energy peak that occurs at 8493 Å in Fig. 9(a) arises from transitions involving $n=2$ conduction levels that become occupied at the highest carrier concentrations. It is unclear at present why the PL observed between 8600 and 8700 Å in samples having lower carrier concentrations (see Fig. 7) disappears in the highly doped samples. No mechanism for this is present in the theory of Lyo and Jones.[19] Measurements on thinner wells ($\sim$100 Å) would help to resolve these questions since there are fewer excited states in this case. However, it is clear that the modulation-doped $Al_yGa_{1-y}As$/$In_xGa_{1-x}As$/GaAs structure provides an ideal system for studying optical properties in the presence of excess free carriers.

V. CONCLUSIONS

A comprehensive review has been given of the photoluminescence properties of GaAs/$In_xGa_{1-x}As$/GaAs SQW's for $0.05<x\leq0.15$ over a range of well thickness ($6\leq L\leq160$ Å). Over the range studied the PL behavior is similar, with both free- and bound-excitonic emission being observed. The PL linewidth as a function of well width has been interpreted in terms of alloy scattering. Detailed information on the properties of the strain-positioned, light-hole excitons has revealed that although the light-hole state is not confined in the $In_xGa_{1-x}As$ well, the excitonic absorption is peaked. The emission properties of modulation-doped $Al_yGa_{1-y}As$/$In_xGa_{1-x}As$/GaAs samples reveal an ideal system for studying the effects of free carriers on the recombination process. A well-defined feature in the PL spectrum of modulation-doped samples can be used to obtain the Fermi-level position, which should provide a valuable characterization of pseudomorphic QW transistor structures.

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