Exciton localization in $In_xGa_{1-x}As$-GaAs coupled quantum-well structures

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Using photoluminescence (PL) and photoluminescence excitation spectroscopy (PLE), we have studied a series of high-quality $In_xGa_{1-x}As$-GaAs coupled multiple-quantum-well structures, grown by molecular-beam epitaxy. The samples are characterized by extremely sharp luminescence lines; the linewidths decreasing from ~4 meV in a sample with wells of 48 Å to ~1 meV in a 14 Å sample. In emission, all the samples exhibit a splitting of the luminescence line into two or three contributions. Splittings of the exciton peaks are also observed in the PLE spectra where, in all cases, the measured energy positions of the heavy-hole exciton creation peaks coincide exactly with the multiple PL lines. We explain our results in terms of small perturbations of either the alloy composition or the $In_xGa_{1-x}As$ thickness which have a dramatic effect on the spatial extent of the wave functions of these "coupled" quantum-well structures. Our calculations demonstrate that by introducing a small degree of randomness in the potential periodicity, the individual heavy-hole states become localized to a few potential wells, giving rise to the multiple exciton peaks observed in our optical data.

INTRODUCTION

The energy positions of the subband states of a semiconductor quantum well (QW) are highly sensitive to changes in the size and shape of the confining potential. Therefore techniques such as photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy which accurately probe the energy states are an excellent means of characterizing this potential and providing quantitative information about, for example, the heterojunction interface. In the GaAs-$Al_xGa_{1-x}As$ QW system Weisbuch and co-workers[1] showed that the luminescence linewidth of excitons reflects the quality of the interfaces and that in good quality samples, the interface roughness can be limited to one atomic monolayer (2.83 Å). More recently, the epitaxial growth of this material system has been further improved and it has been shown tht by interruption of the MBE growth, very large, atomically flat islands can be formed, as evidenced in luminescence by a splitting of the excitonic lines.[2,3] The energy separation between the features observed by these authors corresponds to well-width fluctuations of an integral number of monolayers. In high quality samples grown without interrupts, some workers report PL line splittings equivalent to monolayer changes in layer thickness,[4,5] while others observe a multiplicity of very sharp lines,[6] separated in energy by splittings corresponding to submonolayer fluctuations in well width within the same well. In all cases these investigations have been made on quantum-well structures in which the subband states in adjacent well layers are effectively electronically isolated.

The situation in $In_xGa_{1-x}As$-GaAs QW structures is more complex. In this system the "well" material is the InGaAs alloy, therefore in isolated quantum-well structures variations in the average In composition and/or well width variations can give rise to multiple exciton peaks. Recently we reported the first observation of detailed fine structure in the PL and PLE data of a 14 Å InGaAs-GaAs MQW sample,[7] but could not attempt to make conclusive statements as to the origins of the multiple lines. In this paper we present results on InGaAs-GaAs strained layer MQW samples in which there is significant electronic coupling between adjacent InGaAs layers. We investigate the effects of varying the quantum-well width on the measured energy splittings of the heavy-hole exciton peaks observed in both the PL and PLE spectra. Furthermore, to explain our results we model the effects of introducing small irregularities in the potential periodicity on the spatial extent of the electronic wave functions of these "coupled" structures, demonstrating the effects with calculations applied to the coupled double-quantum-well system.

GROWTH DETAILS AND EXPERIMENTAL TECHNIQUES

The layers were grown in a Varian modular Gen II molecular-beam-epitaxy system on undoped (001) GaAs substrates, rotated at ~20 rpm during growth to ensure lateral uniformity. The substrate temperature was estimated to be 580 °C, which is in the temperature range where significant reevaporation of In occurs. To compensate for this loss an In flux about 1.5 times higher than that normally required to achieve the desired In composition was used. Growth rates for both GaAs and InGaAs were measured using the RHEED oscillation technique on a GaAs monitor slice prior to the growth of each sample. The GaAs growth rate was always set at 1 μm/h and a flux of As_4 was supplied which was just sufficient to ensure an arsenic-stabilized surface for both GaAs and $In_xGa_{1-x}As$. For the specific samples studied here the growth sequence was as follows: (a) a 1-μm GaAs buffer, (b) 5 or 20 periods of InGaAs wells and GaAs barriers, and (c) a GaAs capping layer. The samples were all grown with a nominal In composition of $x=0.12$ and none of the layers was intentionally doped.

We have no evidence that any sample exceeds the critical thickness limit for the relaxation of the compressive strain in the InGaAs and preliminary x-ray analysis[8] of these samples supports this assertion.

PL and PLE spectra were recorded with the samples mounted on the cold finger of a variable temperature (4–300 K), continuous-flow cryostat. The PL measurements were made using an Ar^+ pumped dye (styryl-9) laser, set above the GaAs band gap, at 8060 Å, as the excitation source. The same dye laser provided the tuneable source for the PLE measurements. The luminescence was collected by a double grating monochromator and detected with a cooled GaAs photomultiplier and photon-counting electronics. All the data were recorded at a temperature of ~4 K.

EXPERIMENTAL RESULTS

Figure 1 shows the 4-K PL spectra of four InGaAs-GaAs MQW samples all with nominal In fractions $x=0.12$. The sample with the thickest InGaAs layers is a five period structure with 48-Å wells and 200-Å barriers. The remaining samples all have 20 repeats with 100-Å GaAs barriers and InGaAs wells of either 25, 20, or 14 Å. Infinite superlattice structures with these dimensions would have finite miniband widths associated with both the electron and hole subbands. For such real superlattice systems we calculate $n=1$ electron bandwidths of ~1 meV in a 48–200 Å sample, increasing to 34 meV for a 14-100 Å structure, and $n=1$ heavy-hole bandwidths between <1 and 4.2 meV for the same superlattices. It is clear therefore that we are dealing with truly coupled QW systems. The PL spectrum of each sample is characterized by extremely sharp emission lines, there being either two or three peaks in each spectrum. Repeating the experiment at different points across each sample revealed some variation in the relative intensities of the emission lines but no change to either the spectral positions or the observed splittings. We note a systematic trend to smaller splittings between the peaks as the InGaAs well thickness is decreased. Although all the PL lines are not completely resolved we estimate that the linewidths decrease from ~4 meV in the 48-Å sample to ~1 meV in the 14-Å sample. These are comparable to the best linewidths reported[9,10] and demonstrate the high optical quality of the material. The major factors that contribute to the low-temperature PL linewidth of these undoped QW structures are expected to be alloy broadening and well-width fluctuations. The effect of a fixed variation in the InGaAs layer thickness becomes more significant in narrower wells, because it represents a larger perturbation. On the other hand, alloy broadening effects are reduced as the well width decreases because the exciton wave function extends further into the binary GaAs barrier layers. The observation of a PL line which sharpens with decreasing well thickness (for InGaAs thinner than ~30 Å) has been reported previously.[9] Our data are consistent with the arguments put forward by these authors suggesting that alloy broadening is the dominant mechanism contributing to the PL linewidth.

PLE measurements have been made on all of the samples. In each case two or three spectra were recorded by setting the monochromator to each luminescence peak energy in turn. For illustrative purposes we discuss the data on the 25-Å QW sample, but all our comments about these results apply equally to the other three samples. In Fig. 2 we show again the PL spectrum of the 25-Å sample. Also shown are the PLE spectra recorded with detection energies of (a) 1.4784 eV and (b) 1.4808 eV. With the detection set on the low-energy emission line we can resolve a splitting of the electron to heavy-hole-exciton creation peak of ~1.7 meV. At higher energy (~1.498 eV), in the region of the spectrum where we expect to see the electron to light-hole transition, we again observe a splitting of the excitonic peak. This assignment has been confirmed[11] by subsequent PLE circular polarization experiments, a technique which has been shown[12] to be a useful tool in distinguishing luminescence

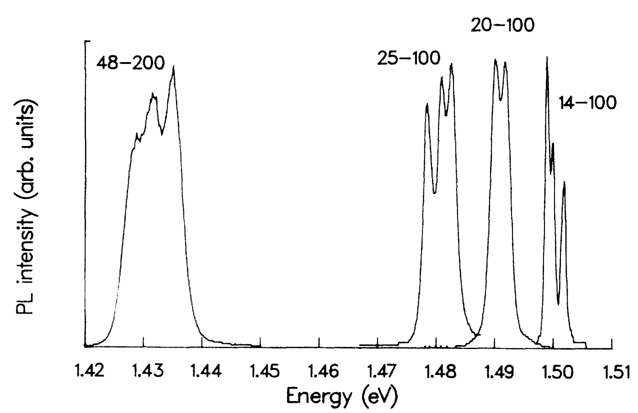

FIG. 1. PL spectra at 4 K from the InGaAs-GaAs coupled MQW samples. The dimensions of the samples are given in Å.

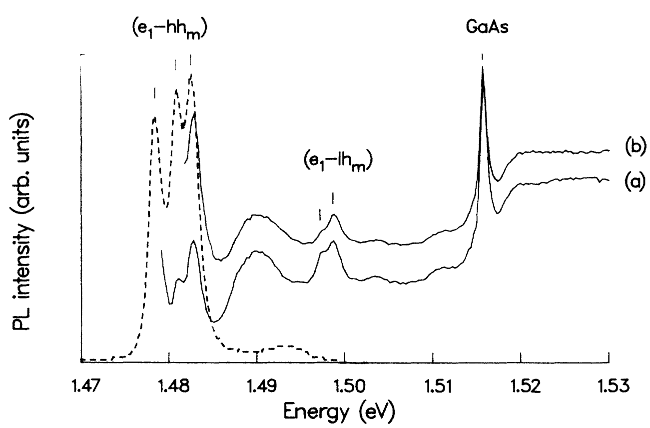

FIG. 2. PL (dashed) and PLE (solid) spectra at 4 K from the 25–100 Å MQW sample. The detection energies used to record the PLE spectra were (a) 1.4784 eV, and (b) 1.4808 eV. The sets of excitonic peaks labeled (e_1-hh_m) and (e_1-lh_m) are due to the recombination of the lowest-energy electron state with the mth localized hole state.

associated with either light or heavy holes. In our experiments the linearly polarized laser light was chopped by an oscillating stress plate to produce alternating $\sigma+$ and $\sigma-$ excitation while detecting changes in one sense of the circularly polarized emission. There are at least two contributions to the light-hole exciton peak, as indicated in the spectra, and perhaps a third unresolved line, broadening the high-energy side. In the region of the excitation spectrum between the heavy- and light-hole excitons, we observe a broad feature with a peak at ~ 1.490 eV. This must, at least in part, be due to the $n=1$ continuum states. However, the absorption strength is clearly enhanced over a simple step-wise density of states and we speculate that this is probably because transitions involving higher-energy electron states also have a contribution in this spectral range. As well as exciton features associated with the quantum wells we also observe a sharp peak at 1.5157 eV which we assign to the creation of GaAs free excitons, predominantly in the buffer and barrier layers.

The two heavy-hole exciton PLE peaks coincide exactly with the luminescence lines at 1.4808 and 1.4825 eV. This argues strongly for the intrinsic nature of both these emission peaks, both corresponding to the creation of free electron to heavy-hole excitons. Moving the detection energy to 1.476 eV, in the tail of the emission, enables us to identify a third heavy-hole exciton in the PLE spectrum. This feature appears as a shoulder at ~ 1.479 eV and we therefore assign the lowest-energy emission peak to a further intrinsic exciton emission. With the detection energy set at 1.4808 eV, shown as curve (b) in Fig. 2, we record essentially the same PLE spectrum. The heavy-hole exciton peak at 1.4825 eV is again observed and there are the same components to the light-hole exciton peak.

ORIGIN OF THE SPLITTINGS

Most observations[2-6] of line splittings in excitonic structure in either PL or PLE are explained in terms of inhomogeneities between or within *isolated* quantum wells. The experimental observations presented in Figs. 1 and 2 are quite different in that they are made on coupled systems. If we consider any coupled MQW sample, we expect the degenerate states of each well to mix and split into multiple, discrete states, each of the resulting states being made up of a linear combination of the exact envelope functions of the isolated wells. Hence the total number of states derived from the original ground state corresponds to the number of periods in the structure. If the sample is structurally "perfect" then all the electronic wave functions of the system are extended over all the quantum wells. In such a situation at low temperatures we expect to see only a single emission line in the luminescence data due to the recombination of electrons and holes that have thermalized to the lowest-energy states. Our explanation of the observed splittings in both the PL and PLE is still in terms of fluctuations in potential but now we believe that these are small random fluctuations in well width and/or indium fraction that are sufficient to localize some of the heavy holes in different regions of the coupled structures, the electron states remaining extended throughout the system. Therefore we assign the multiple exciton peaks to the recombination of the lowest electron state with different perturbed heavy-hole states. The idea of electronic states becoming localized in "imperfect," coupled structures is not a new one—having been investigated theoretically by Lang and Nishi[13] for coupled, double quantum wells using parameters appropriate to electron states in the (InGa)(AsP)-InP system. This work was later extended by Littleton and Camley[14] to include the example of a ten-well coupled system, again using parameters for the quaternary system. In addition, experimental evidence for this effect has recently been reported by Pavesi and co-workers,[15] who made selective-excitation photoluminescence measurements on purposely disordered GaAs-AlGaAs superlattice structures. In each case the authors discussed the problem in the context of the localization-hindering miniband formation at low temperatures and they concluded that slight randomness in the potential periodicity was sufficient to cause an individual electronic state to localize in one or a few of the quantum wells.

We agree with this viewpoint and as an illustration of the phenomenon we show in Fig. 3 effective mass calculations of the first five heavy-hole states of an imperfect 25–100 Å, InGaAs-GaAs, 20-well sample. The sample inhomogeneities we have introduced represent only small imperfections in the structure, which we believe are not unrealistic under our growth conditions. We have randomized the potential by making discrete changes in the well width and depth of 2.5 Å and 3 meV, respectively. These variations correspond to fluctuations in the well width by approximately 1 monolayer and are equivalent to $\sim 1\%$ in 12% change in the In composition. The fluctuations are introduced to half of the wells chosen by a random-number generator such that on average the wells are 25 Å wide and have a mean depth of 40 meV. The heavy-hole mass was assumed to be isotropic through the

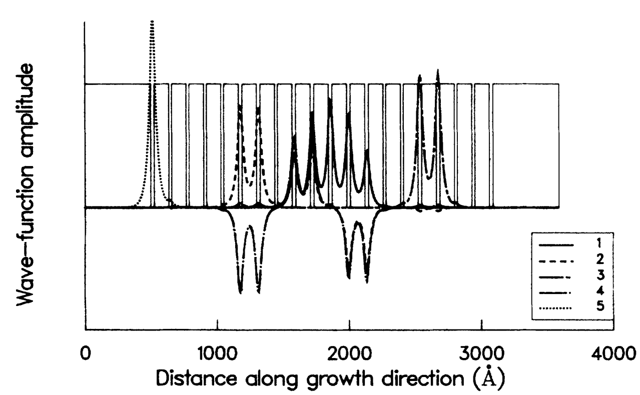

FIG. 3. Calculated heavy-hole wave functions of the 25–100 Å 20-well sample. The potential has been randomized; the well width fluctuates by a monolayer about a mean of 25 Å, and the well depth varies by 3 meV about a mean of 40 meV. Only the first five states are shown.

crystal with a value of $0.34m_0$. Quite dramatic localization of some the states results from these fluctuations; for example, the $n=1$ heavy-hole state resides mostly in the central portion of the wells, whereas, in contrast, the fifth heavy-hole state has become entirely localized near the outermost well.

Within this localization picture discrete lines should occur in the PL spectrum due to the significant overlap of the extended electron states with those hole states which are now localized, and in the PLE we would expect to see exciton creation peaks at the same energies as the emission lines. This is indeed our observation. For example, when the detection energy is set to the lowest energy of the three heavy-hole lines seen in Fig. 2 then the higher-energy components can still be seen. This is because even though the excited hole is localized, the excited electrons have a significant overlap with the created hole states localized at a lower energy, allowing them to contribute to the PL seen at the detection energy. However, the calculations of the heavy-hole wave functions shown in Fig. 3 are only an illustration of the phenomenon and do not allow us to identify which heavy-hole states contribute to the lines observed in the spectra. Such a detailed interpretation would demand an accurate knowledge of the sample inhomogeneities plus a calculation of the oscillator strength of each transition. In this paper we limit ourselves to offering a model consistent with the results and accordingly have labeled the exciton peaks in Fig. 2 as $(e_1-\mathrm{hh}_m)$ and $(e_1-\mathrm{lh}_m)$, where the index m is not identified.

The observed splitting of the light-hole exciton peak in the PLE spectra can also be understood in terms of our localization model. However in this case the situation is complicated by the uncertainty that exists in the literature as to the precise value of the band offsets in this strained system.[16,17] If we assume the fraction of the energy gap differrence between the GaAs and the strained InGaAs electron to heavy hole which apears in the conduction band Q_c to be 0.67, for example, then the light-hole states are type II, being found predominantly in the 100-Å GaAs layers. Alternatively, by choosing a small conduction-band fraction such as $Q_c=0.4$ the light holes are type I and reside in the 25-Å InGaAs layers. We have investigated the extent to which we are able to localize the light-hole states in these two circumstances by producing similar pictures for the light-hole wave functions as shown in Fig. 2 for the heavy holes. We find that if we assume a type-II band alignment $(Q_c \sim 0.67)$ we readily achieve substantial localization of the light-hole states. However, assuming the same degree of randomness, a type-I configuration $(Q_c \sim 0.4)$ produces no signficant localization, the light-hole states remaining extended over all the wells. Therefore, in order to explain our observations within this picture we favor the choice of a band offset ratio which produces a "mixed" system; type I for transitions involving the heavy holes and type II for the light holes. The same choice of offset ratio $(Q_c \sim 0.67)$ also adequately describes the exciton peak positions and the heavy- to light-hole splittings measured in the PLE spectra of all these samples.

As a general comment it is worth noting that the extent to which the electronic wave functions of a superlattice become localized, assuming the same degree of structural imperfection, is dependent not only on the potential barrier height, but also on the layer thicknesses. Lang and Nishi[13] have shown theoretically that the localization tendency can be substantially reduced by decreasing the barrier thickness and/or the confining barrier height. Since we have no reason to assume gross irregularities in the growth of our structures we would not expect to observe multiple luminescence lines in all our coupled MQW samples. The structures used in this study have relatively small effective heavy-hole bandwidths (between <1 and 4.2 meV) and therefore are particularly sensitive to localization effects, due to even slight irregularities in the growth.

The random nature of the fluctuations involved means that it would be difficult and laborious to reproduce exactly the line splittings seen in the PL or PLE. This task is made additionally difficult because, as we have already pointed out, the precise value of the band offsets is an important parameter, since the depth of the potential wells will affect the degree of coupling between the wells and hence the degree of localization (See Ref. 13 for further discussion of this point). However, we can explore whether the localization model is consistent with the trend we see in the line splittings as the InGaAs thickness is reduced. To recapitulate, our observation is that as the InGaAs thickness is reduced the observed splittings of the heavy-hole exciton lines in both PL and PLE become smaller. To illustrate whether this model is capable of describing the observation, we resort to the double quantum well (DQW) as an idealized example. Consider a DQW system of 50-Å InGaAs QW's, separated by a 100-Å barrier of GaAs. If we assume the In fraction is 12%, and the conduction-band offset is 0.67, then using the parameters given in Ref. 18 we calculate the step in the conduction band to be 80 meV and in the valence band as 40 meV. In Table I we show the energy eigenvalues of the two lowest heavy-hole states calculated for the situation where the depth of one well has been perturbed; decreasing it by 3 meV and the width of the wells has been adjusted to maintain the same average nominal well thickness by increasing one well by 2.5 Å and decreasing the other by the same amount. To explore the effect on possible PL and PLE transitions of increased coupling we have made the same calculations for a DQW where the

TABLE I. Calculation of the E_{hh1} and E_{hh2} heavy-hole eigenvalues in the perturbed double quantum well. ΔE is the difference between the two eigenvalues. All energies are in meV and the structure dimensions are in Å.

Ideal structure	E_{hh1}	E_{hh2}	ΔE
50/100/50	14.0	20.7	6.7
25/100/25	25.1	31.2	6.1
14/100/14	32.7	37.5	4.8

well width is first 25 and then 14 Å. The results are also in Table I. Note that upon perturbation the splitting between the heavy-hole states *decreases* as the average well width is decreased. If the splittings in the spectrum are due to recombination of the lowest electron state with the perturbed heavy-hole states then this sort of perturbation which localizes the electronic state will produce a splitting that decreases with decreasing InGaAs thickness as we observe experimentally.

SUMMARY

We have reported optical measurements on InGaAs-GaAs coupled MQW structures. The high quality of the samples is demonstrated by extremely sharp luminescence lines, for example PL linewidths of ~1 meV are measured in a sample with 14-Å wells and 100-Å barriers. All the samples are characterized by a splitting of the PL emission peaks and also by a splitting of both the heavy-hole and light-hole exciton peaks in the PLE spectra. We find that the introduction of a small random interwell fluctuation in the alloy composition and/or the InGaAs well thickness is a sufficient perturbation on the perfect potential periodicity to localize both the heavy-hole and the light-hole wave functions to a few potential wells. This localization model also reproduces our observed trend of a decrease in the measured heavy-hole exciton PL and PLE line splittings with decreasing InGaAs layer thickness.

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