Absorption and photoluminescence of ultrathin pseudomorphic InAs/GaAs quantum wells

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We present absorption data for 2–4-monolayer InAs/GaAs single quantum wells obtained at 77 K using a polarization-based measurement technique. The spectral contribution of the optical loss features arising from bulk GaAs was minimized using the polarization selectivity of absorption in single quantum wells. Based on polarization properties and the results of an envelope-function calculation, the double structure observed in the spectra is attributed to transitions involving confined heavy holes and both confined and unconfined electron states. No evidence of transitions with the participation of confined light holes was found. Well-width fluctuations deduced from the absorption data are a fraction of one monolayer in our samples. The absorption lines were broadened, indicating interface roughness on a scale comparable to the exciton diameter. A single photoluminescence peak overlapping with the lower-energy portion of the fundamental absorption feature was observed in all the samples at 77 K. Although the absorption line of the undoped two-monolayer sample displayed the least broadening, the photoluminescence was red shifted by approximately 70 meV relative to the absorption feature. Presumably, this behavior indicates long-range diffusion of the excitations. Measurements of the temperature dependence and pump-power dependence of the red shift in this sample support this interpretation.

INTRODUCTION

There has been considerable interest in highly strained ($\approx$7% lattice mismatch) InAs/GaAs quantum wells (QW's) both from the fundamental point of view and as a prospective device material.[1] Large lattice mismatch limits pseudomorphic InAs growth to narrow wells. Optical studies[2–7] of QW's up to 5 monolayers (ML) thick have been performed in order to obtain information about their band structure as well as for characterizing the material quality.

We present absorption data for 2-, 3-, and 4-ML InAs/GaAs single QW's (SQW's) obtained using a polarization modulation technique. While photoluminescence (PL) of InAs/GaAs QW's of 1–5 ML thickness has been studied by several groups,[4–7] measurements associated with optical absorption have been reported only for the narrowest wells.[2,3] This is the first optical data other than PL for the InAs/GaAs quantum wells (QW's) wider than $\approx$1 ML.

In recent studies a double peak structure has been observed in $\approx$1-ML multiple quantum wells using photoreflectance (PR) and photoluminescence excitation (PLE) spectroscopy[2,3] and in 1–3-ML SQW's with additional AlGaAs confinement using PL.[4] The lower-energy feature was attributed to transitions between the confined electron and heavy-hole (HH) states. While the workers of Refs. 2 and 3 attributed the higher-energy feature to transitions involving confined electron and light-hole (LH) levels, Lee, Hsieh, and Kolbas[4] have assigned it to transitions between confined electron and unconfined valence states.

We have observed a similar double structure in the absorption spectra. The features have been assigned by comparison of the observed energies and polarization properties to the ones calculated using an envelope-function approximation (EFA). We identify the lowest feature as a transition between the confined electron and heavy-hole states and assign the higher structure to the transition between the confined heavy-hole and unconfined electron states.

The absorption lines were broad indicating interface roughness on a scale comparable to the exciton diameter. The broadening was enhanced as the well width increased. This could be associated with either a change of the scale of the interface roughness or an increase in the roughness.

A single photoluminescence peak a fraction of the linewidth below the center of the lower-energy absorption line was observed in all the samples at low temperatures. The undoped 2-ML sample was exceptional in that it had to be cooled to near 10 K for the photoluminescence to overlap with the absorption line. At higher temperatures the photoluminescence was red shifted by approximately 70 meV relative to the absorption feature. Presumably, this behavior indicates long-range diffusion of the excitations. Measurements of the temperature dependence and pump-power dependence of the red shift in this sample are reported.

SAMPLES AND EXPERIMENTAL RESULTS

Four samples were included in this study. Three samples contained InAs single quantum wells of 2-, 3-, and 4-ML nominal thickness and were not intentionally doped. These QW's were deposited over a 1000-Å GaAs buffer layer epitaxially grown on a semi-insulating GaAs substrate and capped with 500 Å of GaAs. In the fourth

sample, the top 200 Å were doped with Be at $\approx 10^{18}/cm^3$. A diagram of the samples is provided in Fig. 1(a). The samples were grown by molecular-beam epitaxy (MBE) using a technique reported elsewhere.[8] The growth process was monitored by the reflection high-energy electron diffraction (RHEED) oscillations.

Photoluminescence spectra at 77 K have been obtained using a 0.75-m grating spectrometer and conventional lock-in detection techniques. An Ar^+-ion laser (4880-Å line) was used as the pumping source. A Fourier transform spectrometer (FTS) and a He-Ne laser (6328-Å line) were used to study the PL temperature dependence of the undoped 2-ML sample.

Optical absorption measurements have been performed using a FTS. In order to increase the measured QW absorption and to have an option of having a significant fraction of light polarized perpendicular to the QW plane, we used a multiple internal reflection configuration shown in Fig. 1(b). The samples were polished on the substrate side and beveled at 45° at the edge. The light was incident parallel to the QW plane at the beveled edge. After 7–10 internal reflections, the light exited from the opposite edge as shown in Fig. 1(b). Transmission measurements of the undoped 2-ML sample showed a QW feature $\approx 2\%$ masked by a strong background arising in the GaAs substrate and epitaxial layers. The QW signal was completely masked in other samples.

In order to isolate the quantum well absorption features we used a technique based on polarization modulation. The use of this technique is not limited to quantum wells but can be extended to thin absorbing layers of other nature. The ratio of transmitted intensities for incident light polarized parallel ($\parallel$) or perpendicular ($\perp$) to the plane of incidence was obtained. The transmitted intensities are designated $I^{\parallel}$ and $I^{\perp}$, respectively, and the quantity $\ln(I^{\parallel}/I^{\perp})$ will be referred to by the acronym PMA (polarization modulated absorption).

When the light propagates inside the sample [as shown in Fig. 1(b)], interference of the rays incident on the GaAs/air interface with the reflected ones creates a standing wave normal to the interface. The fringe patterns for the two polarizations are spatially displaced because of the polarization dependence of the phase shift upon total internal reflection.[9] Thus the polarization modulation results in a modulation of the optical energy density at the quantum well. This will produce a PMA signal since the thickness of the quantum well is small compared to the fringe spacing. We will show that in addition, due to their different symmetry properties, the transitions involving confined LH and HH states have markedly different PMA signatures and therefore can be easily distinguished.

In contrast, thick regions of GaAs (cap, buffer, and substrate layers) span a large number of fringes. Therefore, for these areas, the field variations due to the fringes are averaged out and the shift of the interference pattern due to the polarization modulation has no effect.

Experimentally measured PMA spectra of the undoped 2-ML QW sample (solid line) and a reference sample consisting of a piece of GaAs substrate with beveled edges (dotted line) are presented in Fig. 2. The polarization-dependent QW features (marked by arrows) are easily identifiable by comparison between the two plots.

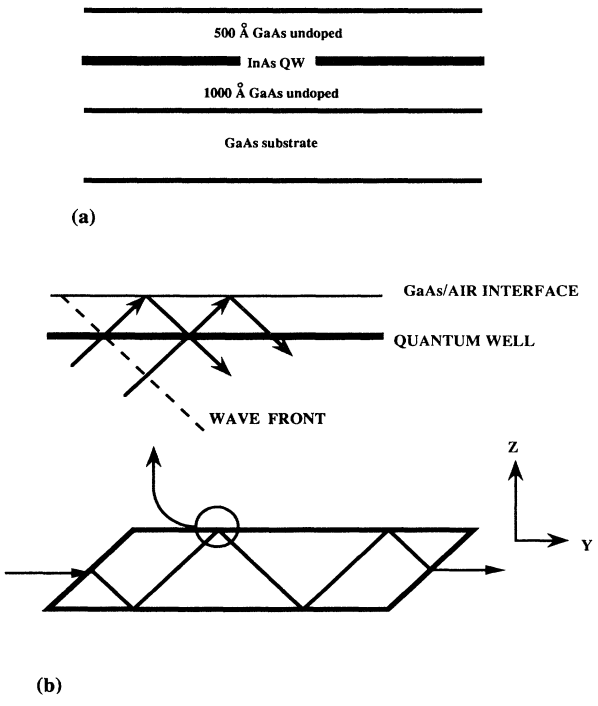

FIG. 1. (a) The sample layout. In the doped 2-ML sample the top 200 Å are Be doped ($\approx 10^{18}/cm^3$). (b) Schematic representation of the light propagation in the sample (lower part) and the light interference at the quantum well plane (upper part).

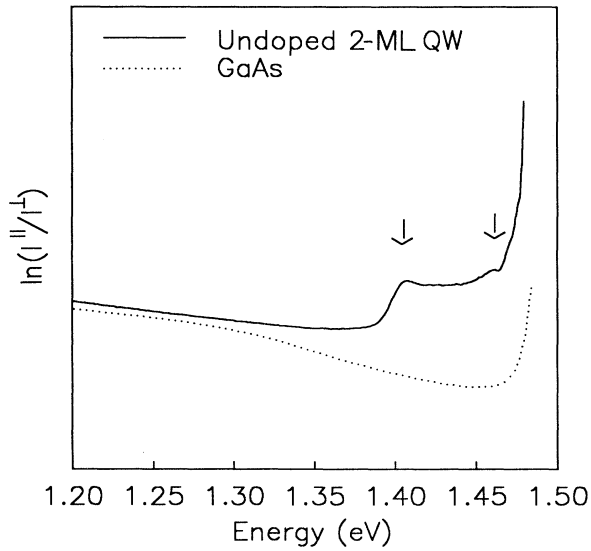

FIG. 2. PMA spectra at 77 K of the undoped 2-ML sample (solid line) and a similarly prepared GaAs reference (dotted line). The observed quantum well transitions are marked by arrows.

Plotting the log of the detected intensity ratio facilitates separation of different attenuation mechanisms since they are multiplicative. Since absorption in unstrained GaAs does not depend on polarization, it will not contribute to PMA spectra. However, the reflection loss occurring when the light enters the sample is polarization dependent and gives rise to a background PMA signal. This background should be almost constant as long as we probe the spectral region below the band gap of GaAs, where the index of refraction is approximately energy independent. The latter condition will change as the GaAs gap is approached. As is apparent in the PMA spectrum observed in the reference sample, in the region of interest the background varies in energy showly enough to be considered as a baseline for the effects due to the QW's (see Fig. 2).

The PMA spectra obtained at 77 K for the undoped 2–4-ML samples are shown by solid lines in Fig. 3 and for the doped 2-ML sample in Fig. 4. PL results at 77 K are also presented along with the PMA results in Figs. 3 and 4 (dotted lines). The PL spectra for 2- and 3-ML wells have been obtained with the pump power density of 4 W/cm^2. The PL spectrum for the 4-ML well was obtained with 100-W/cm^2 pumping.

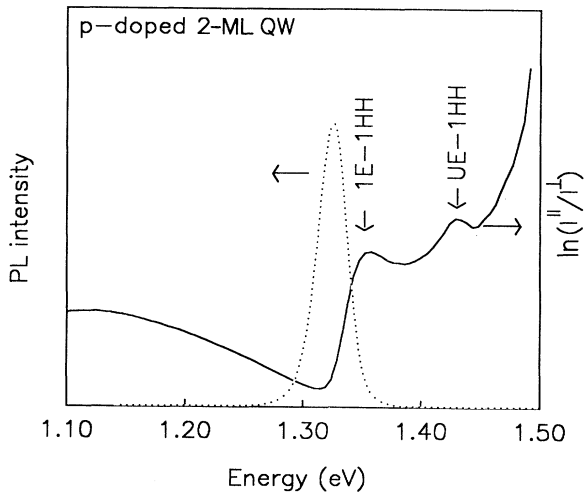

FIG. 4. Photoluminescence (dotted line) and PMA spectra (solid line) at 77 K of the *p*-type doped 2-ML sample. The observed transitions between the confined electron and heavy-hole state (1*E*-1HH) and unconfined electron and confined heavy-hole state (UE-1HH) are marked by arrows.

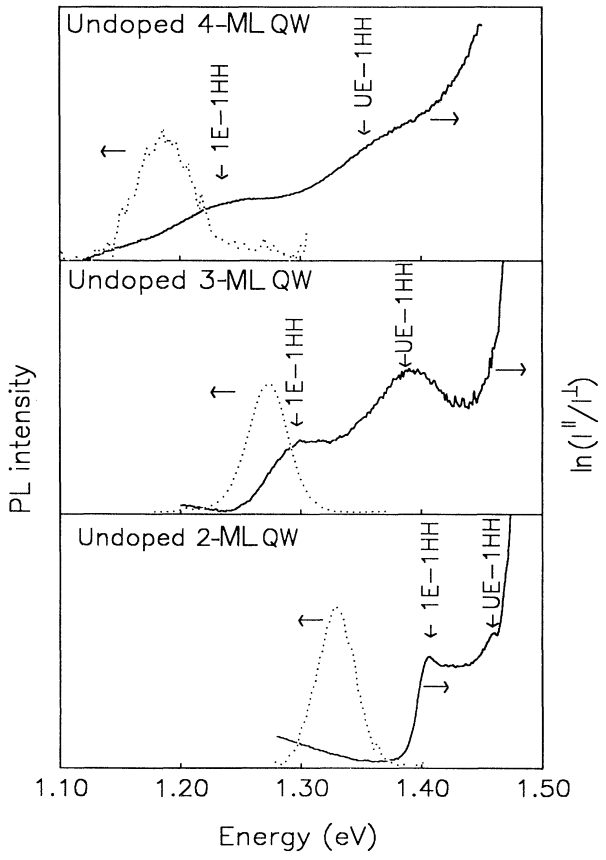

FIG. 3. Photoluminescence (dotted lines) are PMA (solid lines) spectra at 77 K of the undoped 2-, 3-, and 4-ML QW's. The observed transitions between the confined electron and heavy-hole state (1*E*-1HH) and unconfined electron and confined heavy-hole state (UE-1HH) are marked by arrows.

With the exception of the undoped 2-ML sample, the PL spectra at 77 K exhibit a single peak located within the lower-energy wing of the lower absorption feature. The PL peak of the undoped 2-ML sample is red shifted by approximately 70 meV from the absorption peak. We have studied the temperature dependence of the absorption and PL of this sample in the 12–77-K range in order to gain more understanding of the unusually large red shift. The results are presented in Fig. 5. We have also investigated the dependence of the PL peak position on pumping power for this sample. Figure 6 shows PL spec-

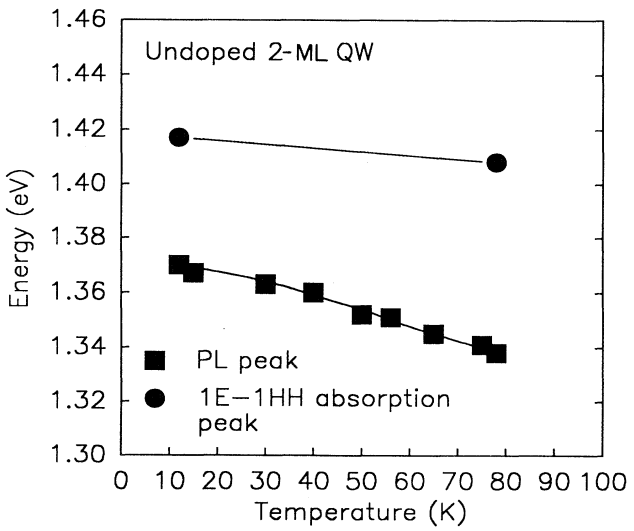

FIG. 5. Photoluminescence spectra at 77 K of the undoped 2-ML QW obtained at various pumping power densities.

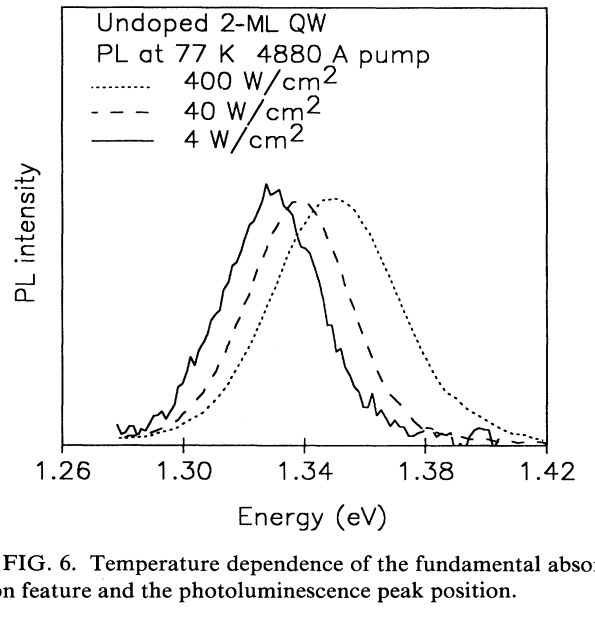

FIG. 6. Temperature dependence of the fundamental absorption feature and the photoluminescence peak position.

tra at 77 K of this sample for pumping power in the 4–400-W/cm^2 range.

DISCUSSION

Absorption

To assist interpretation of the experimental data, we have computed the QW states using an EFA. We have assumed that InAs is pseudomorphic to the surrounding GaAs lattice, and calculated the strain accordingly. Energy gaps and effective masses of InAs have been corrected for strain effects using methods discussed by Pollak.[10] We have adapted the $\mathbf{k}\cdot\mathbf{p}$ treatment of Shuurmans and 't Hooft[11] to the strained QW case by including the strain matrix elements[10] in both the Hamiltonian and the boundary conditions for the mixed light-hole and spin-orbit (s.o.) -split bands. Thus both the $\mathbf{k}\cdot\mathbf{p}$ coupling and strain energy were treated as perturbations of comparable magnitude. The necessary parameters (unstrained band gaps, elastic constants, and deformation potentials) were taken from Ref. 12. We distributed the conduction/HH band discontinuities in the 0.72/0.28 proportion, as proposed by Cingolani *et al.*[3] who arrived at this number by combining the experimental x-ray photoemission data with theoretical corrections of the band positions for strain.

Our calculations indicate the existence of one confined level for heavy holes, light holes, and electrons in all our samples. Thus, in principle, both HH- and LH-related transitions could be present in the spectra. As will be shown below, we are able to discriminate among possible sources of spectral features on the basis of their symmetry properties.

Removal of the degeneracy of LH and HH states by strain or QW confinement leads to strong differences in absorption of radiation polarized parallel and perpendicular to the growth axis.[13] The relative oscillator strength can be derived using Fermi's golden rule for transitions between the p-like LH and HH confined states[11] and s-like conduction (either confined or unconfined) states.

While not affecting the HH wave functions, strain in bulk materials leads to admixing of the s.o. to the LH wave function and may cause a significant alteration of the LH state symmetry properties.[10] However, our EFA calculations indicate that for the 2–4-ML wells the admixture of the s.o. wave function does not exceed 7% and therefore pure LH wave functions[11] were used to derive polarization properties of optical transitions.

The predicted PMA spectra for transitions involving different quantum well states are discussed in the following. In the case of weak absorption the PMA signal $I_{\rm PMA}$ from quantum wells can be written as

$$I_{\rm PMA}\equiv\ln(I^{\parallel})-\ln(I^{\perp})=A^{\perp}-A^{\parallel}\ , \tag{1}$$

where $A^{\parallel}$ and $A^{\perp}$ denote the fraction of the incident power absorbed by the quantum well (QW absorptance) for $\parallel$ and $\perp$ polarizations, respectively.

The absorbed power depends on the polarization for two reasons. First, the strength of the electric field ($\mathbf{E}$) at the well associated with the probe light is determined by the polarization-dependent fringe pattern, as explained in the preceding section. Second, the interaction of the field with the well depends on the field's orientation, as determined by the symmetry properties of the QW states involved in optical transitions.

Using the wave functions of Ref. 11, the absorptances for heavy- and light-hole transitions are proportional to the following functions of the electric field components (E_x,E_y, and E_z) in the space-fixed coordinate system [see Fig. 1(b)]:

$$\begin{aligned} A_{\rm HH}&\propto(E_x^2+E_y^2)/2I_0\ ,\\ A_{\rm LH}&\propto(4E_z^2+E_x^2+E_y^2)/6I_0\ , \end{aligned} \tag{2}$$

where I_0 is the incident-light intensity. With our geometry we then calculate the ratios $A^{\perp}_{\rm HH}/A^{\parallel}_{\rm HH}$ and $A^{\perp}_{\rm LH}/A^{\parallel}_{\rm LH}$. These ratios depend on the photon energy as they are determined by the fringe pattern. Then we express the PMA signal in terms of $A^{\perp}$ and a scaling function $C\equiv(1-A^{\parallel}/A^{\perp})$:

$$\begin{aligned} &\text{HH:}\ \ I_{\rm PMA}=C_{\rm HH}A^{\perp}_{\rm HH}\ ,\\ &\text{LH:}\ \ I_{\rm PMA}=C_{\rm LH}A^{\perp}_{\rm LH}\ . \end{aligned} \tag{3}$$

The magnitudes of $C_{\rm HH}$ and $C_{\rm LH}$ thus found for our samples vary monotonically in the energy range of interest (1.15 eV $\le h\nu\le$ 1.48 eV). The intervals of variation for $C_{\rm LH}$ and $C_{\rm HH}$ are listed in Table I. Thus the absorption features can be reconstructed from the PMA spectra. Since $C_{\rm HH}$ and $C_{\rm LH}$ change slowly, relatively sharp QW features can be observed directly in the PMA spectra.

These results also provide a means of identifying the confined hole states involved in optical transitions: according to Table I, HH transitions provide a positive contribution to the PMA signal, while the LH absorption results in a negative signal. We have no model describing the symmetry of the unconfined hole states and have

TABLE I. Range of variation of C_{LH} and C_{HH} for $1.15 \leq h\nu \leq 1.48$ eV.

C_{LH}	C_{HH}
−0.68 to −0.29	0.85 to 0.75

made no predictions regarding polarization properties of the transitions involving these states.

All spectra exhibit two positive features (such as in Fig. 2) which is consistent with either transitions involving confined HH or unconfined valence states. Thus, in contrast to previous works,[2,3] we do not observe transitions between confined electron and LH states (1*E*-1LH). The lowest-energy feature is ascribed to the transition between the bound electron and HH states (1*E*-1HH). The higher-energy feature can be attributed to either transitions involving the bound HH and unbound electron states (UE-1HH) or the unbound valence and bound conduction states (1*E*-UH). A comparison of the experimental data and calculated energy levels suggests that the assignment of the higher-energy feature to the UE-1HH rather than 1*E*-UH transition provides better agreement. Transitions between confined and unconfined states have been observed previously in uncoupled multiple QW's and SQW's.[14]

To summarize our results, in Fig. 7 we have plotted the positions of the absorption features (squares) for 2–4-ML undoped QW's versus the well width. Also plotted in Fig. 7 are the results of our EFA calculation for all possible QW transitions discussed above. The dotted, solid, dashed, and dash-dotted lines represent UE-1HH, 1*E*-1HH, 1*E*-1LH, and 1*E*-UH transitions, respectively. The agreement between the experimental and calculated energies for both 1*E*-1HH and UE-1HH transitions is good and could be further improved by adjusting the well widths and the band offset used in our calculations. However, we use the envelope function calculation as a qualitative guide only.

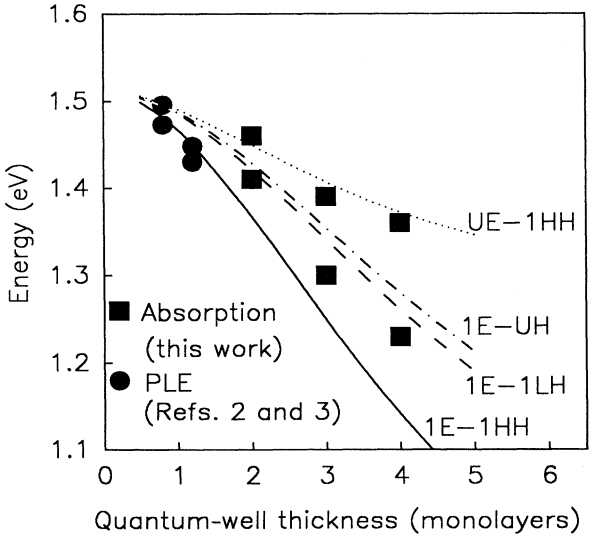

FIG. 7. Experimental and theoretical transition energies for InAs/GaAs QW's vs the well width. Experimental results of this work (squares) and Refs. 2 and 3 (circles) are presented. Curves show theoretical results for all possible transitions in this system.

For comparison, we have also included in Fig. 7 the results of previous PLE experiments.[2,3] A striking feature of this data is that the splitting between the observed transitions decreases with an increase in the well width. This is contrary to the theoretical prediction that the splitting should increase which is also obeyed by our data as well as the results of Ref. 4. The source of this disagreement is not clear at the moment.

There is considerable discrepancy between the transition energies observed for the undoped and *p* type doped 2-ML samples. A deviation of the doped sample from the nominal thickness can explain this disagreement. Both absorption structures observed for this sample fit in the experimental data in Fig. 7 if a well width of approximately 2.5 ML is assumed. As was pointed out in Ref. 2, observation of such noninteger well widths can be made possible by interface roughness on a scale finer than the exciton diameter.

Our discussion of the absorption line shapes is confined to the 1*E*-1HH feature since a simple theoretical description exists for transitions between the two bound states.[15] If there is little inhomogeneous broadening, one expects to observe line shapes mimicking the steplike two-dimensional joint density of states with a peak at the low-energy edge due to excitonic effects. The line shape observed in the undoped 2-ML sample (see Fig. 3) is close to this ideal one. Since the transition energy is sensitive to the layer thickness, interface roughness on a scale comparable to the exciton diameter ($\approx$250 Å) broadens out the absorption line shape.[16,17] In our samples the inhomogeneous broadening increases with increasing well width (see Fig. 3) which may result from the increased interface roughness or change of scale of the roughness.[18] The greatest broadening (approximately 60 meV) is observed in the 4-ML sample; it corresponds to well-width fluctuations on the scale of the exciton diameter of a fraction of one monolayer.

Photoluminescence

Figure 3 shows that while the absorption features broaden significantly with the increased well thickness, the PL linewidth does not display a similar trend. Another surprising feature of the PL spectra (Figs. 3 and 4) is that although the undoped 2-ML QW has the sharpest absorption features, it displays the largest PL red shift at 77 K (approximately 70 meV below the absorption line).

This behavior can be explained if the carrier diffusion is taken into account. In photoluminescence, the generated carriers are captured by the well and then diffuse[16,17,19] (possibly as neutral excitonic particles[19]) with subsequent radiative recombination. Thus the PL line shape depends not only on the distribution of the ex-

citon states[20] but also on the exciton mobility within this distribution. For example, it was found that the mobility edge determines the PL peak position in disordered ternary semiconductors.[21]

For the undoped 2-ML sample, the extent of the PL peak suggests that a tail extending below 1.38 eV exists in the exciton distribution and that the excitons are able to diffuse to the states below the absorption edge so that recombination happens primarily in the tail. We believe that a similar tail exists in the exciton distribution of other samples, although we cannot observe it directly in the PMA spectra because of the background. The smaller PL red shift in these samples points to the inability of excitons to diffuse to the tail states before recombining. This could be caused by either a sharp drop of the exciton mobility in the lower portion of the absorption line[16] or shorter nonradiative recombination lifetimes.

Since the undoped 2-ML QW displays the narrowest absorption line, it probably has the most ordered interfaces and thus all excitons within the absorption peak could behave like free particles with high mobility, similar to the high-quality QW's of Ref. 22. But, even if this is not the case and the mobility edge exists within the absorption line (as was found for the quantum wells used in Ref. 16), mobility of the excitons in the localized regime may be enhanced in comparison with other samples since this is the thinnest well in this study.[17]

We have measured the temperature dependence of the separation between the absorption and PL line in the undoped 2-ML QW. As Fig. 5 indicates, this separation decreases from 72 meV at 77 K to 48 meV at 12 K. Thus at least part of the shift can be explained by the exciton mobility in the localized regime due to the thermal activation. Takagahara[17] have found that contribution of this mechanism decreases significantly as the sample is cooled.

Since the mobility of localized excitons is lower than in the free state, a relatively modest increase in the pumping power should fill up the exciton distribution tail and affect both position and broadening of the PL line. In Fig. 6 we present PL peaks obtained from the undoped 2-ML sample using 4880-Å excitation with the pump power density varying from 4 to 400 W/cm^2. All peaks are normalized to the same height. In agreement with the prediction, with increased pumping power the peak position shifts to higher energies by as much as 20 meV and the line broadening is increased.

CONCLUSIONS

We have studied undoped InAs/GaAs SQW's of 2-, 3-, and 4-ML width and one modulation-doped (p-type) 2-ML sample. Absorption spectra have been measured using a polarization-based technique. This is an optical study of InAs/GaAs QW's in this well-width range by a method other than PL. Based on their predicted energies and polarization properties, the absorption features attributed to transitions from heavy-hole confined levels to both confined and unconfined conduction states were observed. No evidence for the light-hole-related transitions have been found. The experimental dependence of the energy of these transitions on the well thickness follows the trend predicted by an envelope-function calculation. The extent of the absorption line broadening suggests that the size of the well-width fluctuations is a fraction of one monolayer in our samples.

A single photoluminescence peak was observed in all the samples. Spectra of the undoped 2-ML sample displayed an unusually large red shift of the photoluminescence in comparison to the fundamental absorption structure. This behavior was explained in terms of exciton diffusion, a model supported by measurements of temperature and excitation-power dependence of the photoluminescence.

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