

Origin of sharp lines in photoluminescence emission from submonolayers of InAs in GaAs

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Very sharp and intense low-temperature photoluminescence (PL) emission is reported from submonolayers of InAs in GaAs grown by flow modulation epitaxy at 50 Torr using trimethylindium and tertiarybutylarsine. InAs coverages from 0.25 to 1.0 monolayers were systematically studied using low temperature PL, and photoluminescence excitation spectroscopy and temperature-dependent, time-resolved PL. The observed PL linewidths and energies are satisfactorily explained within a two-dimensional (2D) quantum well picture over all coverages. In these ultrathin structures we see confirmation of existing models which predict a smooth transition of the PL linewidth to zero in the limit of extremely thin wells. The possibility of weak 0D localization of excitons by thickness variations in the submonolayer samples is discussed. [S0163-1829(97)06407-2]

I. INTRODUCTION

Considerable attention has been recently devoted to monolayer and submonolayer InAs/GaAs heterostructures. The emission from single monolayers (ML) of InAs in GaAs has been observed for some time in samples grown by atomic layer epitaxy (ALE), molecular-beam epitaxy (MBE), and metalorganic chemical vapor deposition (MOCVD).¹⁻³ Such structures show very intense photoluminescence (PL) with extremely high quantum efficiencies, suggesting possible applications for laser devices.⁴ Recently there have been several reports of highly efficient luminescence from submonolayers of InAs deposited in GaAs by molecular-beam epitaxy. Physically this system is very interesting because it represents the limit of decreasing well width for 2D quantum well structures. The question of how the detailed in-plane distribution of In in these layers affects the optical properties of excitons in these extremely thin wells is an active area of research. Recently there have been several suggestions that very thin submonolayer quantum wells grown by MBE exhibit evidence of 0D or 1D localization.⁵⁻⁸ One outstanding issue is whether the in-plane distribution of InAs in submonolayer wells can lead to a localization strong enough to modify the exciton energies appreciably from the value expected for a uniform $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer with the appropriate composition as recently proposed by Wang *et al.*⁶ An alternative picture for these structures proposed by Alonso, Ilg, and Ploog and recently supported by others⁸ is that for low In coverage the in-plane transport of excitons becomes inhibited because of islanding of In within the plane. In this paper we report on recent results obtained on well-characterized submonolayer InAs structures grown by flow modulation epitaxy (FME). From an analysis of PL linewidths we conclude that the submonolayer samples consist of monolayer islands of InAs with roughly a 2 nm size. Since the exciton radius is much larger than this, the sample behaves essentially like a very thin quantum well. The question of whether or not in-plane exciton transport is suppressed as

postulated by Alonso, Ilg, and Ploog⁷ is difficult to verify. We show here that in the present samples, PL excitation (PLE) and temperature-dependent time-resolved PL data are consistent with purely 2D unhindered in-plane exciton transport, however, some degree of localization cannot be ruled out. In contrast to previous works,^{5,6} we see no evidence that the exciton energy spectrum is appreciably modified by 0D or 1D confinement.

II. EXPERIMENT

The InAs/GaAs quantum wells were grown in a vertical MOCVD reactor designed to permit *in situ* characterization techniques such as reflectance difference spectroscopy. Details of the growth apparatus were given in detail elsewhere.⁹ Single monolayer and submonolayer InAs/GaAs structures were grown simultaneously on (001) GaAs substrates with nominally exact and 2° miscut toward (110) orientations. Atomic force microscope (AFM) scans of the exact substrates revealed regular terraces with some fingering of the terraces out along the miscut direction. The residual miscut of the nominally exact substrates was determined by AFM to be around 0.1°. The investigated samples were grown on a 2 μm -thick high-quality buffer layer grown at 600 °C by conventional MOCVD using triethylgallium and tertiarybutylarsine (TBA). PL from this material showed very sharp bulk excitonic luminescence. Monolayers or submonolayers of InAs were deposited at 390 °C using trimethylindium (TMI) and TBA in a four-step cycle consisting of a 3-s hydrogen purge followed by a TMI pulse of 0.5–3 s, a 3-s hydrogen pulse, and a 6 s TBA pulse. Following the InAs layer, approximately 1.3 ML of GaAs was grown by FME at 390 °C in order to reduce In segregation. The temperature was then ramped up to 500 °C in order to desorb any floating layer of InAs. This technique has been shown to be effective in the MBE growth of similar structures at comparable temperatures¹⁰ and gives us good agreement with the expected PL emission for one ML of InAs in GaAs.

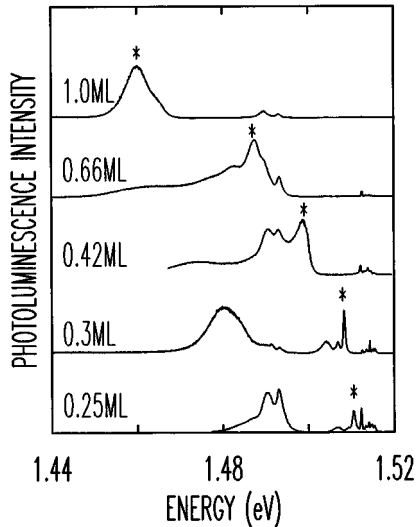


FIG. 1. PL emission from monolayer and submonolayer quantum wells of InAs in GaAs as a function of In coverage. Asterisks indicate the position of the In-related excitonic emission.

High resolution x-ray-diffraction measurements were performed on the samples in order to establish the number of incorporated In ML. A BEDE 150 diffractometer equipped with a four crystal Ge channel cut monochromator operated in the (220) reflection was used in conjunction with a 2 kW generator using a copper target.

For the optical measurements, the samples were mounted strain free in a He-pumped cryostat. The PL was excited using the 514-nm line of an Ar-ion laser. The signal was dispersed by a double grating 0.85-m spectrometer and detected by a GaAs cathode photomultiplier in photon counting mode. Photoluminescence excitation (PLE) spectra were obtained using a tunable Ti-sapphire pumped by an Ar-ion laser. The time-resolved measurements were performed using a pulsed Ti-sapphire laser. The 2-ps pulses were tuned at the GaAs free-exciton resonance. The PL signal was detected with a GaAs photomultiplier using the time-correlated photon counting technique.

III. RESULTS

Figure 1 shows the PL from samples grown with various InAs layer thicknesses inserted in GaAs. No significant difference was observed between the PL properties of samples grown on nominally exact or miscut (001) substrates. In the case of the 1-ML and 0.3-ML samples, the total deposited In was determined from x-ray-diffraction measurements combined with dynamical scattering theory simulations as reported by Brandt *et al.*¹⁰ Details of our simulations will be presented elsewhere. Such determinations are believed to be fairly reliable as far as the total dosage of InAs but the simulations are not sensitive to the exact location of the inserted In. In other words the simulations cannot distinguish between one ML of InAs or two ML of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ since both of these situations result in the same net displacement of the epilayer and substrate GaAs lattices. Nevertheless, for a given total In dosage, the PL emission is sensitive to the distribution. For the other samples shown in Fig. 1, the incorporated In was estimated from the known In exposure

TABLE I. Structural parameters, PL and PLE energies of the InAs submonolayer quantum wells investigated in this work.

| Sample | InAs coverage (ML) | E_{PL} (eV) | E_{PLE} (eV) | PL FWHM (meV) |
|--------|--------------------|----------------------|-----------------------|---------------|
| S1 | 1 | 1.460 | 1.464 | 7 |
| S2 | 0.66 ^a | 1.482 | <i>n/a</i> | 6 |
| S3 | 0.42 ^a | 1.499 | 1.500 | 3 |
| S4 | 0.3 | 1.507 | 1.507 | 0.7 |
| S5 | 0.25 ^a | 1.511 | 1.511 | 1 |

^aThese values were determined from the known pulse length of TMI.

time combined with the known x-ray values for the 1-ML and 0.3-ML samples. The observed PL energy of 1460 meV for the 1-ML sample agrees with the generally accepted value for 1-ML quantum wells of InAs in GaAs, which has been calculated theoretically and confirmed experimentally by several groups. In addition, high-resolution transmission electron microscopy on samples with this emission wavelength showed roughly 1 ML of InAs localized to a single atomic plane.¹⁰

As the In incorporation is reduced, the energy of the In related emission varies smoothly towards higher energy, eventually merging with the GaAs bulk luminescence for coverages below 0.25 meV. This smooth variation is reasonable if we assume that the submonolayer In wells are composed of monolayer high InAs islands with an extent and separation much smaller than the exciton radius. Thus one does not expect to see in this system discrete peaks corresponding to one or two ML high regions as observed in other systems with larger effective masses (e.g., GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$). At intermediate In coverages (0.4–0.65) the submonolayer luminescence is obscured somewhat by the free-to-bound and donor-acceptor pair peaks of the GaAs barrier layers at 1.493 and 1.490 eV. For coverages below 0.4 ML we observe very sharp luminescence features, with a linewidth as low as 0.7 meV for the 0.3-ML sample. The energies and linewidths of the various PL transitions are given in Table I.

For low coverages, e.g., 0.3 ML we observe in addition to the sharp principal peaks labeled 1, weaker broader peaks labeled 2 and 3. These lower-energy peaks saturate with increasing laser power, indicating that they are of a different physical origin from peak 1. Reflectance measurements on the 0.3-ML sample only show a peak at position 1, indicating that this is due to a free-excitonic transition, and hinting that the other two peaks represent bound-state transitions, possibly donor or acceptor bound excitons. The temperature dependence of these lines adds further support to this hypothesis as shown in Fig. 2. Peaks 2 and 3 quickly disappear as the temperature is increased while peak 1 remains sharp and visible up to 80 K. The laser intensity was increased for the higher-temperature spectra in order to compensate for the drastic reduction in signal. Beyond 80 K, both peak 1 and the GaAs free-exciton-polariton luminescence disappear due to thermal dissociation. The shift to lower energy with increasing temperature is just due to the reduction in band gap with increasing temperature. It is important to note that the very narrow linewidth of peak 1 is not necessarily evidence for

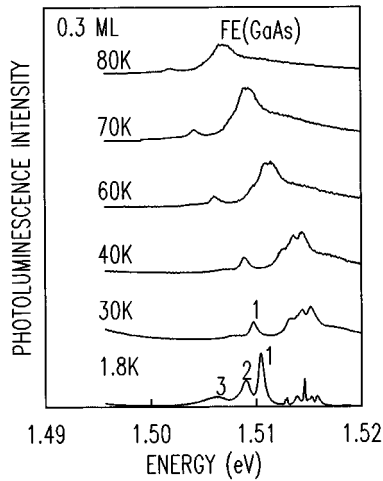


FIG. 2. Temperature dependence of the 0.3-ML In submonolayer PL showing the disappearance of lines 2 and 3 with increasing temperature.

the suppression of thermal broadening by 0D or 1D localization, since the selection rules for exciton polaritons permit emission only at the photon momentum of $\sim k=0$. Thus the observed linewidth does not represent the true distribution of exciton energies, but only those which emit light. In addition, the fact that peak 1 disappears at roughly the same temperature indicates that there is not a significant enhancement in the exciton binding energy for this transition compared with bulk GaAs. The data do not rule out the possibility however of a weaker form of localization such as the suppression of in-plane exciton transport postulated by Alonso, Ilg, and Ploog⁷ which would also tend to suppress thermal broadening.

In addition to the excitonic peaks shown in the 0.3-ML sample, there is also a weaker impurity band at approximately 1.48 eV shifted down by approximately the bulk acceptor binding energy. This peak saturates with increasing pump power and we attribute it to free-to-bound or donor-acceptor pair emission in the vicinity of the In submonolayer. A similar band, shifted down in energy by the same amount is observed in the other samples.

PLE measurements have been used in support of claims of 0D or 1D confinement in submonolayers of InAs in GaAs. In Fig. 3 we compare the PL and PLE data for the 1 ML sample. The PL spectrum shows a broad peak with a linewidth of 7 meV as well as weaker excitonic features from bulk GaAs. In PLE, we observe a Stokes shift of roughly 4 meV of the PLE to higher energy as is usual for narrow 2D quantum wells with interface roughness. In addition, we see another resonance at higher energy which is consistent with the electron-to-light-hole transition observed by other groups for single ML InAs.

As the In coverage is reduced we see a steady reduction in exciton linewidth together with a reduction in the Stokes shift to roughly zero by 0.3 ML (Table 1). In Fig. 4 we present the PLE and PL results for the 0.3 ML sample. Figure 4(a) shows the result of monitoring the PL at the low-energy impurity emission at roughly 1480 meV shown in Fig. 1. This PL band is associated with donor-acceptor pair or free-to-bound emission in the vicinity of the InAs submonolayer. Figures 4(b) and 4(c) show, respectively, the

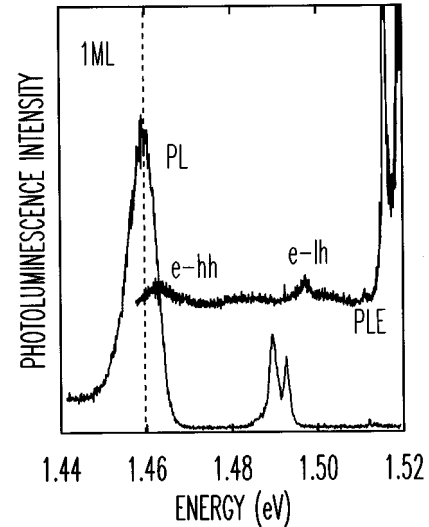


FIG. 3. Photoluminescence excitation spectra for the 1-ML sample showing the significant Stokes shift with respect to PL.

PLE spectrum obtained while monitoring at the impurity related peaks 2 and 3 indicated in Fig. 4(e). Figure 4(d) shows the effect of monitoring the PL at the low-energy side of peak 1. Sharp features corresponding to the e -lh transition as well as GaAs excitonic transitions are seen in all three cases. No significant shifts in the PLE energies of the light- or heavy-hole transitions were observed as a result of monitoring at these different wavelengths. The linewidth of the e -lh transition is approximately 0.25 meV.

IV. DISCUSSION

Two models have been recently proposed to explain luminescence emission from submonolayers (~ 0.3 ML) of InAs in GaAs.^{5,7} Wang *et al.*⁶ and Belousov *et al.*⁵ have reported very sharp ~ 0.5 meV linewidth PL emission from

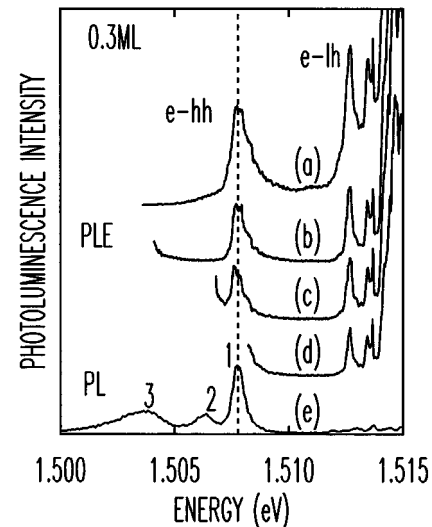


FIG. 4. Photoluminescence excitation spectra for 0.3-ML sample obtained at various monitoring wavelengths. (a) monitoring submonolayer impurity band, (b) monitoring transition 3, (c) monitoring transition 2, (d) monitoring low-energy side of transition 1. (e) shows the nonselective PL emission for comparison.

nominal 0.3-ML InAs layers deposited by MBE. In Ref. 7, 1D quantum confinement effects are invoked to explain the energies of the observed submonolayer PL emission for 0.3 ML InAs layers. This assessment is based on a discrepancy between their observed emission energies and their calculation assuming ultrathin InAs quantum wells. The observed discrepancy is relatively small however, and easily within the range of systematic errors which could arise from the x-ray method used to determine InAs coverage. In addition the choice of exciton radii and band offsets can affect the shape of the fit. A final source of discrepancy is the known presence of some level of In segregation in these types of structures. However, even if one assumes that 1D confinement somehow increases the exciton binding energy for low In coverage as claimed in a related paper by Wang *et al.*,⁶ it is very difficult to imagine how InAs islands could be so uniform as to give the sharp PL features observed. Certainly, in the case of other self-organized systems, very broad linewidths are usually reported unless lateral patterning is used to reduce the observed island size distribution.¹¹

Alonso, Ilg, and Ploog have observed strong PL features at In coverages around 0.3 ML in samples grown by MBE at temperatures comparable to the present study.⁷ The lines in that study were considerably broader with widths of around 4 meV for 0.3-ML samples. In their model, for samples with coverages below 0.4 ML, the excitons are basically frozen at islands of InAs with a diameter and separation substantially lower than the exciton diameter. In that model the exciton energies are unaffected by confinement, the only effect of confinement is a freezing of the center-of-mass motion. The key piece of evidence proposed in favor of their interpretation is the observation of a small 0.5-meV redshift of the PLE compared with PL when monitoring on the low-energy side of the PL transition. This is rather weak evidence for localization however, and could be caused by other inhomogeneous broadening mechanisms, such as large scale variations in the well width, so it is of interest to find other techniques for verifying or disproving this model.

Li *et al.*⁸ recently observed time-resolved photoluminescence emission from submonolayers and monolayers of InAs in GaAs and interpreted their data along the lines of Alonso, Ilg, and Ploog. They observed a difference in the lifetime between 1.0, 0.5, and 0.3 ML samples, however the trends were not systematic. On the one hand, they saw an increase in lifetime going from 1.0 to 0.5 ML which they attributed to in-plane localization. On the other hand, the lifetime of the 0.3-ML layer was the same as the 1-ML sample and this was claimed to arise from competition from nonradiative centers. In view of the very preliminary nature of their data, confirmation of appreciable localization for submonolayer wells cannot be conclusively inferred from that work.

Before addressing the issue of 0D or 1D confinement, we first show that the energy spectra and PL linewidths are basically described by a simple 2D excitonic picture. As a starting point we have performed a simple effective-mass envelope calculation of PL emission energies as a function of well width assuming that the well width varies uniformly to zero and including strain via the continuum elastic constants. The effect of exciton binding energy is not included. The results are shown in Fig. 5 assuming conduction and valence-band offsets of 624 and 416 meV, respectively. The

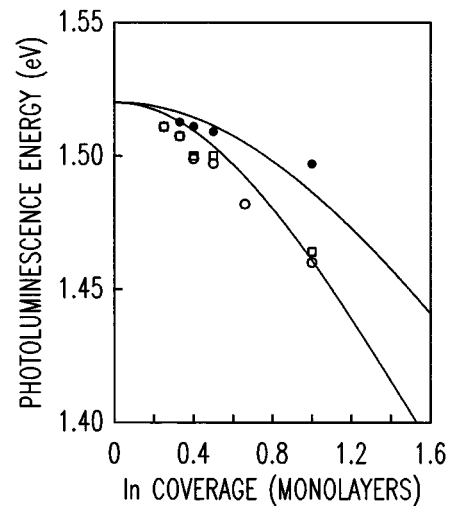


FIG. 5. PL emission energies (circles), PLE heavy-hole energies (squares), and PLE light-hole energies (solid circles) emission energies plotted as a function of In coverage. The solid line represents the result of the simple effective-mass envelope calculation.

agreement is reasonable considering the primitive nature of the model, which is clearly not expected to be very accurate for such thin layers. Our experimental points agree reasonably well with the experimental points of other MBE grown samples.⁵⁻⁸

Physically, an InAs submonolayer can be visualized as either a series of InAs monolayer islands punctured by GaAs or as a uniform $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer with the appropriate composition. Singh and Bajaj¹² have presented a model for linewidth fluctuations for quantum wells. Two main contributions to the linewidth are possible and include monolayer thickness fluctuations, and alloy fluctuations of the quantum well material. For very thin quantum wells, the effects of interface roughness are known to produce appreciable broadening. This broadening leads to the usual observation of a Stokes redshift of PL relative to PLE due to the migration of excitons to lower-energy regions of the well in PL. The linewidth is predicted to increase with decreasing well width up to a certain point due to the combined effects of interface roughness and alloy broadening. However in the limit of zero thickness, models for both mechanisms predict a tendency toward zero linewidth. This is due to the increasingly bulklike behavior of excitons in very narrow wells, whose energy must eventually converge to bulk GaAs in the limit of zero thickness as the perturbation of the InAs layer becomes less and less.

In Fig. 6 we plot our measured PL linewidths as a function of In coverage. As predicted by models of both interface and alloy broadening we see a smooth reduction of the linewidth to zero in the limit of zero coverage. The dashed line in Fig. 6 is based on a very simple estimate of the effect of alloy broadening assuming a single monolayer of $\text{In}_x\text{Ga}_{1-x}\text{As}$ with $0 < x < 1$. In this simple estimate we assume that the exciton bound to the submonolayer plane has the bulk GaAs exciton radius of 14 nm,¹³ a reasonable assumption since the exciton penetrates deeply into the bulk GaAs. Assuming Gaussian statistics, the fluctuation in the number of In atoms N in this area is just \sqrt{N} . This fluctuation in N can be correlated with an energy variation from Fig. 6

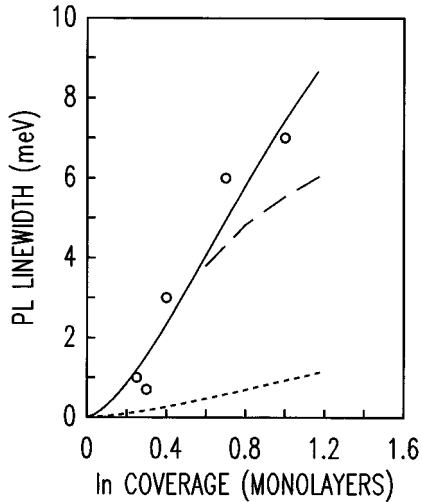


FIG. 6. Measured linewidths as a function of In coverage. The solid line represents theoretical interface broadening assuming InAs islands of radius 1.0 nm. The dashed line is the fitted data from MB samples of Patané *et al.* (Ref. 14). The dotted line represents theoretical linewidths for a uniform 1 ML $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer with no interface broadening.

which gives a simple estimate of the linewidth shown by the solid line. The above results indicate that alloy broadening is not the dominant source of broadening in our samples if we assume a uniform $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer. If the In is distributed randomly, then some additional interface roughness or possibly, In segregation is required to model the data accurately.

In an alternate physical picture of the submonolayer wells they can be treated as islands of monolayer InAs surrounded by GaAs intrusions. If we suppose that the island size is much smaller than the exciton radius, the exciton energies will be essentially the same as for the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer at comparable coverage. If the island size is assumed to be uniform for the sake of simplicity, the linewidth can be estimated by counting the number of islands within an exciton radius. Assuming the placement of islands is random then the same simple method used above gives us an estimate of the linewidth. The solid line in Fig. 6 shows the result assuming an island radius of 1.0 nm. A full analysis of linewidths is beyond the scope of this paper and in any case irrelevant considering that it is extremely difficult to determine the exact microscopic configuration of the In in the layer and thereby compare theory with experiment. Whatever the broadening mechanism, it is clear that the linewidths converge to zero in the limit of low coverage as expected. It is interesting to observe that Patané *et al.* have reported linewidth results for InAs wells over the thickness range 0.8–1.2 ML, and the observed linewidths agree within 1 meV with our observations over that range.¹⁴

PLE results were used by Alonso, Ilg, and Ploog to argue for suppression of excitonic transport for submonolayer wells.⁷ In the present work, the data of Fig. 4 show no evidence of a Stokes shift for coverages below 0.4 ML. If excitonic transport in the plane were not inhibited, one would at first sight expect a Stokes shift of the order of half the PL linewidth of 0.7 meV, whereas none is observed. The Stokes shift would be of the order of around 0.3 meV. However, for the measurement temperature of $T=1.8$ K thermalization

TABLE II. Time-resolved PL data.

| Sample S1 temperature (K) | Lifetime (ns) | Sample S4 temperature (K) | Lifetime (ns) | Intensity |
|------------------------------|------------------|------------------------------|------------------|-----------|
| 6 | 0.8 | 6 | 0.6 | 1 |
| 20 | 1.0 | 20 | 0.4 | 0.27 |
| 40 | 2.1 | 30 | 0.4 | 0.073 |
| 60 | 0.7 | 40 | 0.5 | 0.014 |

would prevent any bias towards the lower-energy regions, and hence no Stokes shift would be expected with or without excitonic motion.

Temperature-dependent time-resolved PL measurements were performed on the 1.0 and 0.3 ML samples in order to determine whether there is any evidence of appreciable in-plane excitonic transport. At a temperature of 6 K the lifetime of the 0.3-ML sample is somewhat shorter than that of the GaAs layer. Since there are many factors that can alter PL emission lifetimes, little can be inferred from such a result. A general feature of radiative emission from 2D wells is that the lifetime should increase with temperature due to the increasing population of states with crystal momentum other than $k=0$. Since these are forbidden by polariton selection rules from recombining, the recombination rate decreases with increasing temperature. This is clearly observed for the 1.0-ML sample as shown in Table II. This confirms the physical picture of in-plane exciton motion indicated by the Stokes shift between PL and PLE for this sample (Fig. 3). In contrast the 0.3-ML sample is essentially temperature independent as one would expect at first sight for the case of excitons with suppressed in-plane mobility, for which $k=0$ regardless of temperature. However, this conclusion cannot be inferred from this data for the following reason. Table II shows that the intensity of the 0.3-ML sample is strongly decreasing with increasing temperature. The origin of this behavior can be realized by noting that for this In composition, the submonolayer exciton is only bound by about 10-meV relative to the free exciton. For temperatures above 10 K there is therefore a strong probability of quenching of the quantum well excitons to free bulk excitons. A detailed study of the temperature dependence of the time-resolved data will be presented elsewhere, however, the high-temperature activation energy for the intensity data is consistent with the observed spectroscopic binding energy of 10 meV. Thus the lack of a significant change in the PL lifetime at high temperatures merely confirms that the dominant recombination channel is the relatively fast free-exciton recombination. The overall reduction in 6-K lifetime between the 1.0-ML sample and the 0.3-ML sample can be understood as due to an increase in the electron-hole overlap perpendicular to the quantum well. For the 1.0-ML well, the hole is much more strongly perturbed than the electron by the monolayer insertion. The strength of this perturbation decreases significantly as the coverage is reduced to 0.3 ML.

The question of the exact distribution of the In within the submonolayer regions cannot be addressed in detail in this study. Bressler-Hill *et al.* and other have observed wirelike islands of InAs on GaAs by scanning tunneling microscopy.¹⁵ Whether or not such features survive the pro-

cess of burial under GaAs in that form as suggested by Wang *et al.* has yet to be verified. Given the large degree of segregation of In following GaAs overgrowth reported by several authors, it is unlikely that such features retain the exact form indicated in the STM studies. In any case, to the first order it makes no difference to the exciton spectrum what the exact in-plane distribution of In is within limits. If the exciton radius is much larger than the length scale of the islands or wirelike regions then the perturbations on the energies should be small compared with an identical coverage of $\text{In}_x\text{Ga}_{1-x}\text{As}$. For low coverages the exciton wave function has very little overlap with the submonolayer plane, and is thus not expected to be strongly affected by the exact distribution of In.

V. CONCLUSION

In summary, we have studied the optical properties of very sharp and intense PL emission lines from submonolayers of InAs in GaAs grown by flow modulation epitaxy. The linewidths of these very narrow wells were observed to ap-

proach very low values as expected in the limit of extremely thin wells. PL linewidth measurements as a function of thickness indicate that the In is clumped in the form of monolayer high islands with a radius of roughly 1.0 nm. The data indicate that the exciton radius is much larger than the cluster spacing and that the PL emission energies are essentially those of a 2D ultrathin quantum well. We cannot rule out the possibility of 0D localization effects in our samples, but conclude that they are not necessary to explain the present data.

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¹³This value is calculated using $a = \epsilon a_0 m_0 / \mu_0$. Where a_0 is the hydrogen atom Bohr radius, $\epsilon = 12.5$ is the dielectric constant of GaAs, $\mu_0 = 0.048 m_0$ is the reduced effective mass of the exciton as quoted in S. B. Nam, D. C. Reynolds, C. W. Litton, R. J. Almassy, T. C. Collins, and C. M. Wolfe, *Phys. Rev. B* **13**, 761 (1976).

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