Photoluminescence and stimulated emission from monolayer-thick pseudomorphic InAs single-quantum-well heterostructures

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Spontaneous- and stimulated-emission spectra from a series of $Al_{0.3}Ga_{0.7}As$/GaAs/InAs separate-confinement strained single-quantum-well heterostructures are demonstrated for well widths as thin as 1 monolayer (3 Å). These undoped samples, grown by molecular-beam epitaxy, are the thinnest single quantum wells ever reported to support stimulated emission. Continuous-wave (cw) laser thresholds at 77 K are generally quite low (0.78 kW/cm^2) despite the fact that the single quantum wells are undoped and of dimensions (L_z) which were previously thought to be too small to effectively collect excess carriers ($L_z \ll$ scattering path length). A simple model based on the spatial extent of the wave function, rather than the well width, is proposed to explain the experimental results. Also a simple modified square-well model with strain-induced band-gap correction is found to predict the experimentally measured energy levels of ultrathin quantum wells.

I. INTRODUCTION

In recent years, quantum-well-heterostructure lasers have attracted a significant amount of interest because of their superior characteristics compared to conventional double-heterostructure lasers.[1-3] The main factors contributing to this superiority are modified density of state and quantized energy levels. Also, considerable research effort has been devoted to the growth, characterization, and device applications of strained-layer semiconductor heterostructures.[4] The ability to grow defect-free crystals of this type allows the fabrication of superlattice and quantum-well heterostructures from a much wider variety of constituent materials than strict adherence to lattice-matching constraints would allow. This significantly extends the range of materials and properties available in layered semiconductors (e.g., InAs/GaAs heterostructures). High-quality strained single quantum wells can be grown by different methods, provided that the thickness of the strained layers are kept small enough to avoid the generation of dislocations.[5,6] For such layers, the lattice mismatch is taken up by elastic strain. The critical thickness, h_c, for dislocation generation has been found to be approximately inversely proportional to the In content.[5]

In this work we are concerned with the carrier collection necessary to support stimulated emission in ultrathin strained-layer quantum-well heterostructures. Previously we suggested that strain may improve the carrier-collection efficiency for very thin (50 Å) pseudomorphic quantum wells due to increased scattering at the interface.[7] However, recent results[8-10] and our current work show that the spatial extent of the wave function (not the well width) is more important than strain effects when considering the capture of hot carriers.

Early work on $Al_xGa_{1-x}As$/GaAs single quantum wells indicated that the carrier-collection process in a thin ($L_z \ll 100$ Å) undoped well becomes ineffective and thus cannot sustain stimulated emission.[11,12] As the width of the quantum well approaches or becomes smaller than the scattering-path length, the carriers jump over the well rather than scattering (losing energy) and being captured by the quantum well. Heavy doping[13] and coupling of multiple quantum wells[1] were proposed and shown to provide increased efficiency in carrier collection. Also, separate-confinement heterostructures have been shown to improve laser performance by creating a reservoir of carriers surrounding the quantum well so that the carriers make multiple passes over (through) the well increasing the overall collection efficiency.[14] Theoretical models have also been presented which confirm that as the quantum-well thickness approaches the scattering-path length the collection efficiency decreases.[12]

Recent experiments[10,15] have demonstrated that single quantum wells of $Al_xGa_{1-x}As$/GaAs as thin as 3 monolayers (ML) and GaAs/InAs as thin as 2 ML are efficient enough collectors of carriers to support cw stimulated emission at 77 K. The success of these results does not require the use of multiple, coupled quantum wells, heavy doping, or graded band-gap confining layers, although the separate-confinement structure was used. Successful operation of both lattice-matched and strained-layer ultrathin quantum wells suggests that something more fundamental than strain accounts for the successful laser operation of single, undoped quantum wells with dimensions much less than the carrier scattering length. In this paper we demonstrate that previous models for carrier collection are in qualitative agreement with these new experimental results if the well width is replaced by the spatial extent of the wave function. We also suggest a simple model for the calculation of bound states in the case of ultrathin strained quantum wells.

II. EXPERIMENTAL PROCEDURE

The pseudomorphic GaAs/InAs quantum wells described in this work were grown by molecular-beam epitaxy (MBE) (Varian 360) on Si-doped (100)-oriented GaAs substrates. The growth (0.5 ML/sec) of the laser structures proceeded as follows. First, a 6000-Å-thick $Al_{0.3}Ga_{0.7}As$ cladding layer was deposited with the substrate held at 640 °C. The active region, consisting of an InAs single quantum well of width L_z, sandwiched between two 500-Å-thick GaAs confining layers, was then grown at a substrate temperature of 550 °C, with 2-sec growth interruptions at both interfaces. Finally, a second 6000-Å-thick $Al_{0.3}Ga_{0.7}As$ cladding layer was grown at 640 °C to complete the symmetrical structures. All the layers in these laser structures are undoped. A series of consecutively grown samples with $L_z = 1$, 2, 3, and 4 ML, and a control sample without an InAs quantum well (0 ML) were grown.

Sample preparation for standard photoluminescence experiments consists of cleaving small rectangular sections from the MBE-grown wafers and mounting them on a copper heat sink using indium. Additional preparation for photopumped laser samples consisted of selectively removing the substrate from the epitaxial layers by mechanical polishing and selective chemical etching, cleaving the remaining film into rectangular platelets 20–100 μm in width, and pressing the platelets into indium under a sapphire window.[16] The excitation source was an argon-ion laser operated continuously or pulsed ($\lambda = 5145$ Å, 8-nsec pulses at 3.8 MHz). Luminescence from the samples was collected and analyzed using a 0.5-m spectrometer and an S-1 photomultiplier.

In this work, a simple, modified square-well model with a strain-corrected band gap is used to calculate the bound states of the quantum wells. The boundary conditions $\Psi_- = \Psi_+$ and $\Psi'_- = \Psi'_+$ were used. A concentration-dependent band offset[17–20] was used to fit the experimental data to the modified square-well model. Some interesting properties for the bound-state calculation of these pseudomorphic ultrathin quantum wells are briefly discussed in Sec. III.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Standard photoluminescence data

77-K photoluminescence spectra from the structures (1, 2, 3, and 4 ML and control sample) for low excitation levels (< 100 W/cm^2) are shown in Fig. 1. The single strong peak from the control sample (0 ML) indicates the high quality of the GaAs confining layer. In the 1-ML single-quantum-well spectrum there are two strong narrow peaks, one from a quantum-well bound-state transition, probably the $n = 1$ electron to unbound light or heavy hole,[19–21] and one from the GaAs confining layers, respectively. Spectra from the 2- and 3-ML single-quantum-well samples exhibit three peaks. The major strong peak corresponds to the $n = 1$ electron to unbound light-hole or unbound heavy-hole transition, and the peak at lower energy corresponds to $n = 1$ electron to $n = 1$ heavy-hole transition. The strong transition is exhibited because the light holes are confined in the GaAs confining layers by the AlGaAs barriers and the heavy holes are confined in the InAs quantum-well region. This results because the degenerate valence band splits in the case of a biaxially compressed quantum-well heterostructure. Thus, all the light holes (and for other reasons many of heavy holes) remain in the GaAs layer, and the $n = 1$ electron to unbound-hole transition is dominant in

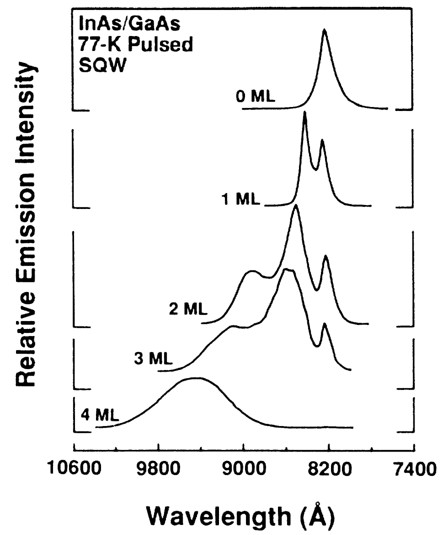

FIG. 1. 77-K standard photoluminescence spectra for all four of the strained single-quantum-well samples and a control sample.

TABLE I. Comparison between experimental photoluminescence peak data and the calculated transition energies based on two different models. e, hh, and uh denote electron, heavy hole, and unbound hole, respectively.

			Transition energy (eV)	
		1 ML	2 ML	3 ML
Square well	e-hh	1.497	1.463	1.414
($\Delta E_c = 0.85$)[a]	e-uh	1.502	1.486	1.460
Modified				
square well	e-hh	1.468	1.393	1.361
($\Delta E_c = 0.6$				
to 0.775)[b]	e-uh	1.497	1.474	1.431
	e-hh		1.391	1.365
		1.468[c]	1.415[c]	1.390[c]
Experimental				
	e-uh	1.477	1.461	1.444
		1.491[d]	1.456[d]	1.434[d]

[a] Fixed.
[b] Concentration dependent.
[c] From Gerard *et al.*, Ref. 24.
[d] From Taira *et al.*, Ref. 23.

the 1-ML sample. As the well width increases, the luminescence from the electron to heavy-hole transition becomes stronger and is observed in the spectra from the 2 and 3 ML samples (Fig. 1). This occurs because (1) the binding energy (relative to the top of the valence-band well) increases producing a larger heavy-hole population in the well and (2) the reduction in the size of the $n=1$ electron wave function reduces the overlap with the unbound-hole wave functions. The reduction in the GaAs confining-layer luminescence (8240 Å) with increasing L_z is the result of the combined effects of collection, retention, and recombination of the carriers in the quantum well. The relative importance of these processes is discussed later. In the case of the 4-ML single quantum well, the photoluminescence peak is shifted toward lower energy and is very broad and weak. We attribute these undesirable characteristics of the 4-ML (13 Å) sample to the generation of misfit dislocations or the onset of three-dimensional growth. This is consistent with the critical thickness of 15 Å predicted using the model of Matthew and Blakeslee[5] and also is in good agreement with the previous data of Andersson *et al.*[22] Finally, we note that the higher-energy emission, electron to unbound hole, is consistent with the data reported by Taira *et al.*[23] while the lower-energy emission electron to heavy hole is consistent with Gerard and Marzin.[24] A summary of these data appear in Table I and Fig. 2.

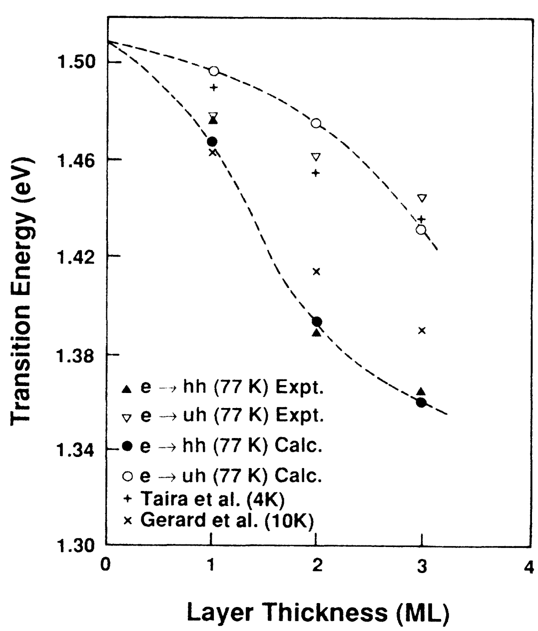

FIG. 2. Experimental photoluminescence peak data and calculated dependence (dashed line) of the transition energies vs InAs well thickness. Corresponding data from Taira *et al.* (Ref. 23) and Gerard *et al.* (Ref. 24) are shown by pluses and crosses, respectively.

B. Modified square-well model for a pseudomorphic ultrathin quantum well

In biaxially compressed $In_xGa_{1-x}As$ the z component of the hole effective mass is significantly smaller for the split-off or light-hole ($j_z=\pm\frac{1}{2}$) valence subband than for the heavy-hole ($j_z=\pm\frac{3}{2}$) one. For this reason, the observed decrease in energy difference between the $n=1$ electron to unbound hole and the $n=1$ electron to $n=1$ heavy-hole lines, with a reduction in L_z, could indicate a nonconfinement of the holes belonging to the strain-split-off valence subband.[20] Hence, the valence-band offset in $In_xGa_{1-x}As$/GaAs heterostructures should be smaller than the strain-induced valence-band splitting as shown in Fig. 3. The x dependence of the alloy band gap $E_g(x)$ is given by the expression[25]

$$E_g(x)=E_g(0)-1.47x+0.375x^2 \text{ eV} . \tag{1}$$

The strain-induced band-gap shift for a biaxially compressed layer is

$$\delta E_g(\varepsilon)=2a_G\left[1-\frac{C_{12}}{C_{11}}\right]\varepsilon-b\left[1+2\frac{C_{12}}{C_{11}}\right]\varepsilon . \tag{2}$$

The deformation potential a_G which represents the band-gap change per unit hydrostatic dilation, is related to the easily measured hydrostatic pressure coefficient of the gap as

$$a_G=-\tfrac{1}{3}(C_{11}+2C_{12})\left.\frac{\partial E_g}{\partial P}\right|_{P=0} . \tag{3}$$

Typical values of a_G are on the order of -8 eV for GaAs and -3 eV for InAs.[25] Typical values of the valence-band deformation potential, b, are on the order of -2 eV for GaAs and InAs. The lattice mismatch is $\varepsilon(0.07)$, and

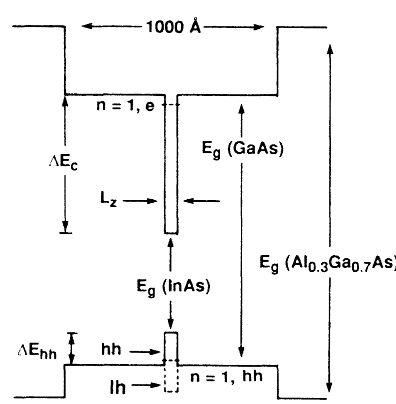

FIG. 3. Energy-band configuration of a strained InAs/GaAs/$Al_xGa_{1-x}As$ separate confinement single-quantum-well structure. In the InAs layer the valence band is split into a $j_z=\pm\frac{3}{2}$ (heavy-hole) and a $j_z=\pm\frac{1}{2}$ (light-hole) band. The lowest energy for a heavy hole is the $n=1$ heavy-hole bound state in the InAs well and the lowest energy for a light hole is the band edge of the GaAs confining layer.

C_{ij} are the elastic constants. Using a simple square-well model, the equations above, and assuming a conduction-band discontinuity of 0.85,[18,20] we can calculate the spectral position of transitions for $In_xGa_{1-x}As$ pseudomorphic quantum wells. The calculated data for the $n=1$ electron to $n=1$ heavy-hole and the $n=1$ electron to unbound-hole transitions for all investigated samples are given in Table I. These values are not a good fit to the experimental data, especially the $n=1$ electron to $n=1$ heavy-hole transition, because we did not account for the strained GaAs layer at the interfaces. For thicker (>20 Å), less highly strained InGaAs/GaAs quantum wells, the simple quantum-well model with the assumption of strain only in the InGaAs has been successful. However, for a monolayer-thick InAs/GaAs quantum well, the interface must play a much more important role, and we expect the strain in the adjacent GaAs layers to be important. The InAs layers would be biaxially compressed and the GaAs layer biaxially dilated[25,26] such that the lattice constant of an ultrathin strained quantum well would be some value intermediate to the InAs and the GaAs bulk lattice constant. If the GaAs layers on each side of InAs quantum well are under tensile strain due to the compressive strain in the InAs, a more accurate calculation of the bound states can be obtained: The 1-, 2-, and 3-ML-thick InAs quantum-well band structures on the left-hand side of Fig. 4 will then be equivalent to those on the right-hand side. Using this modified model, a variable band-gap offset and the intermediate lattice constant a_i,[25] where

$$a_i = 5.653 + 0.405x + 0.81x\frac{(5.38-0.85x)}{(11.88-3.55x)}\ , \qquad (4)$$

the bound states of all the investigated samples were recalculated and are shown in Table I. These values are in good agreement with the experimental data with an error of less than 20 meV or 1.35%. For the 1-ML sample, the sharp peak at 8410 Å corresponds to the $n=1$ electron to unbound-hole transition, but we do not observe the $n=1$ electron to $n=1$ heavy-hole transition or we are unable to resolve it. For the 2- and 3-ML samples, we observe that the sharp, strong peaks correspond to $n=1$ electron to unbound-hole[21] transitions and the weaker, broader peaks correspond to the $n=1$ electron to $n=1$ heavy-hole transitions, and match the calculated values within a small error (<13 meV, or 0.9% error). The good match to the experimental data is due in part to the fact that for ultrathin quantum wells the binding energy (relative to the confining layer) is insensitive to variations of the effective mass in the well and insensitive to the depth of the well. The larger full width at half maximum of the photoluminescence peaks for the 2- and 3-ML quantum wells in Fig. 1 may be ascribed to the onset of misfit-dislocation generation or the onset of three-dimensional growth.

C. Photopumped laser operation

The 77-K stimulated emission spectra from three different samples are shown and compared with the control sample in Fig. 5. Note that stimulated emission occurs at the calculated ($n=1$ electron to unbound-hole) particle transition at a reasonable power density in spite of the fact that the wells are much smaller than the carrier scattering-path length, and carrier collection is expected to be quite poor. Obviously, from Fig. 5, the laser action occurs at the bound quantum-well state rather than in the GaAs confining layers. Note that the excitation power density for the 1-ML sample is smaller than the control sample. This decrease is attributed to the fact that the InAs quantum well can efficiently capture and

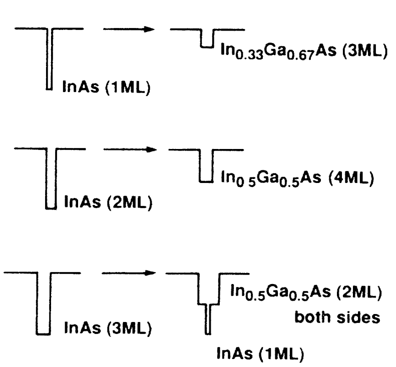

FIG. 4. Modified square-well model. 1-, 2-, and 3-ML-thick InAs quantum-well band configurations on the left are equivalent to those on the right if we assume that the GaAs layers on both sides of InAs quantum well are under tensile strain of the same magnitude as the compressive strain in the InAs.

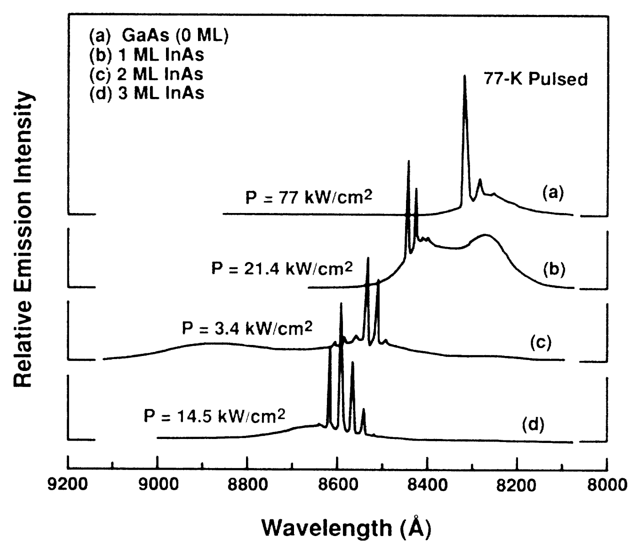

FIG. 5. 77-K stimulated emission spectra of three different InAs single quantum-well samples and a control sample. The observed spectra occur at the calculated $n=1$ electron to unbound-hole particle-transition energies. These are the thinnest quantum-well structures to exhibit stimulated emission.

recombine enough carriers to support stimulated emission before population inversion occurs in the GaAs confining layers. The 2-ML sample is also an efficient collection and recombination site with the added advantage that the binding energy (relative to the GaAs) is large enough to suppress the emission of carriers from the well (discussed later) and consequently suppress the GaAs emission. The rise in power density for the 3-ML sample is attributed to the fact that the critical thickness is approximately 4–5 ML, and the threshold is expected to rise as the well dimension approaches the critical thickness. These are the thinnest GaAs/InAs quantum wells to exhibit stimulated emission yet reported. A continuous-wave laser threshold as low as 780 W/cm^2 was measured at 77 K from the 2-ML sample, as shown in Fig. 6. These results provide clear evidence that photoexcited carriers are effectively collected and low-threshold laser operation is possible using ultrathin single quantum wells.

Efficient carrier collection was achieved in an ultrathin single quantum well without heavy doping, coupling of multiple quantum wells, or a graded index structure. A partial explanation of the unexpected performance of these samples can be found in Ref. 14, where the influence of the separate confinement structure on carrier collection is described. That is, photoexcited carriers are first trapped near the quantum well within the 1000-Å active region, which is a combination of a waveguide and a reservoir. They then transverse back and forth across the well until scattered to lower energy in the well and collected. Thus, the overall capture probability can be increased, but the single-pass capture probability is not necessarily changed.

The separate-confinement heterostructure alone cannot account for the laser action from the ultrathin quantum wells in this work. First, the carrier lifetime in the reservoir is very short when stimulated emission occurs in the GaAs confining layers. Secondly, other work has shown that good carrier collection and laser action can be obtained in the ultrathin quantum wells without a separate confinement structure.[8,9,27] We believe that the spatial extent of the wave function and not the well width is the more fundamental length parameter when evaluating carrier collection. For example, if an electron or hole scatters outside the well, it can lose energy and be captured by the well if the wave function extends outside the well.

Given the simple square-well potential, the wave functions can be determined if the well width, well depth (including strain-corrected band gap for the strained layer), and effective masses are known. The full width at half maximum (FWHM) of the probability density functions for the bound-electron and heavy-hole states of a GaAs/InAs quantum well are shown in Fig. 7. The FWHM of the probability density at first decreases as the well width decreases, but then begins to increase for very thin well widths. For a wide quantum well, a carrier must scatter within the well to couple to the bound-state wave functions. Decreasing the well width decreases the wave function, and hence, decreases the probability that an inelastic scattering-capture event will occur within this smaller region of space. However, for sufficiently thin wells, further reducing the well width increases the

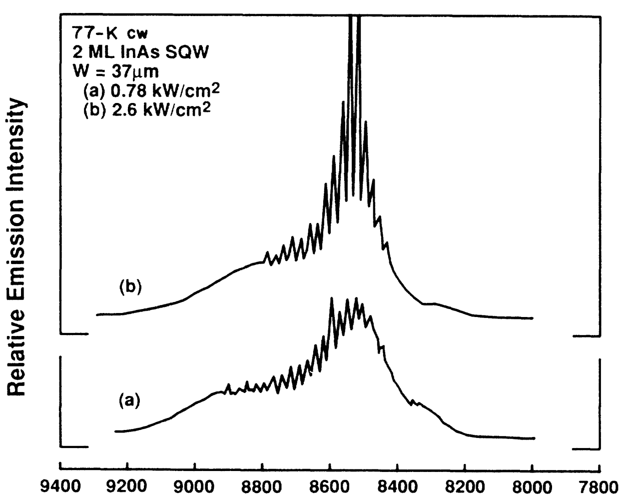

FIG. 6. 77-K cw stimulated-emission spectra from a 2-ML InAs/GaAs single-quantum-well structure. Note that the threshold of 0.78 kW/cm^2 is comparable to the threshold from other, much thicker ($L_z >$ scattering-path length) quantum wells.

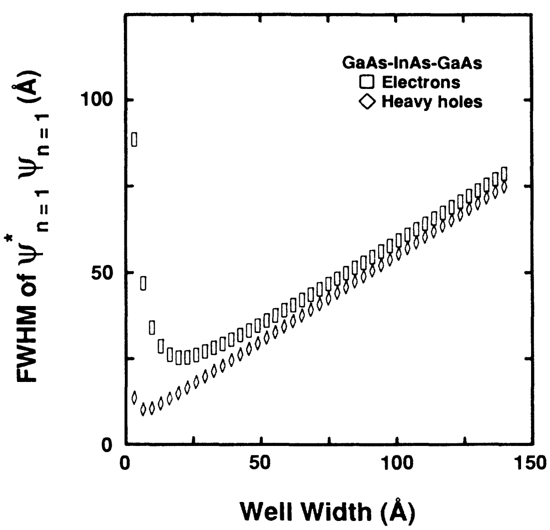

FIG. 7. Calculated full width at half maximum (FWHM) of the $n=1$ bound-electron and heavy-hole probability densities as a function of well width (in increments of 1 ML). The FWHM of the probability density at first decreases with well width, but then increases for very thin wells. Note that the electron and heavy-hole curves have a different functional dependence because of the difference in effective mass and the relative splitting of the conduction- and valence-band discontinuities.

spatial extent of the wave function, and the probability of capture increases because the interaction distance (volume) increases. This analysis reinforces our findings that the spatial extent of the wave function should be considered rather than the well width of the quantum well when evaluating the capture of carriers by a quantum well.

Carrier collection is a necessary, but not sufficient condition for laser action to occur. The recombination depends on both the occupancy of the states and the matrix element for the transition. Note in Fig. 7 that the shape of the heavy-hole curve differs from the electron curve in that the heavy-hole wave function continues to decrease with decreasing well width while the electron wave function begins to increase with decreasing well width. This difference in behavior of the wave functions will reduce the oscillator strength of the electron to heavy-hole optical transitions (by a few percent) and change the relative collection efficiency of the electrons versus the holes as a function of the well width. If only the collection efficiency is considered, the ratio of quantum well to barrier emission should be higher for the 1-ML samples than the 2- or 3-ML samples (see Fig. 5), which is not observed. However, if the escape of carriers from the well and the relative oscillator strengths are taken into account, then the experimental data can be explained as follows. The oscillator strengths differ by only a few percent. However, the binding energies (relative to the GaAs confining layers) of the electrons and heavy holes are 5 and 7 meV, 21 and 25 meV, 48 and 47 meV, for the 1-, 2, and 3-ML samples, respectively. At 77 K, $kT = 6.6$ meV, which is substantial compared with the electron binding energy in the 1-ML quantum well. Hence, a larger amount of recombination from the GaAs barriers is expected from 1-ML sample relative to the 2- and 3-ML samples as observed in Fig. 5. Hence, the experimental results agree qualitatively with the sample wave-function model and demonstrate that reducing the well thickness can improve the chances of obtaining quantum-well laser emission in the ultrathin well region.

IV. CONCLUSIONS

We have demonstrated photoluminescence and photopumped laser operation in undoped, monolayer-thick, single-quantum-well heterostructures grown by molecular-beam epitaxy including the thinnest, a 1-ML, single-quantum-well heterostructure ever reported to support stimulated emission. Good carrier collection and laser operation were achieved without multiple wells, heavy doping, or a graded band-gap structure, although a separate-confinement structure was used. The experimental results demonstrate that carrier collection can be efficient in ultrathin quantum-well heterostructures.

A simple model based on the spatial extent of the wave function rather than the well width was presented. This model qualitatively explains the differences between the experimental observations and the expected laser performance of ultrathin quantum wells in previous studies. Replacing the well width with the spatial extent of the wave function appears to rectify the discrepancies without changing the basic physics of the models or contradicting any previously reported data. Finally, a simple modified square-well model was developed for ultrathin strained-layer quantum-well heterostructures which correctly predicted the experimentally measured optical transition energies.

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