

Hole-state reversal and the role of residual strain in (In,Ga)As-GaAs superlattices

Karen J. Moore, Geoffrey Duggan, Gerke Th. Jaarsma,* Paul F. Fewster, and Karl Woodbridge
Philips Research Laboratories, Redhill, Surrey RH1 5HA, England

R. J. Nicholas

Clarendon Laboratory, Parks Road, Oxford OX1 3PU, England

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We use photoluminescence excitation (PLE) spectroscopy to investigate the electronic properties of $\text{In}_{0.10}\text{Ga}_{0.90}\text{As-GaAs}$ superlattice (SL) structures in which compressive and tensile strain are incorporated simultaneously in alternate layers. This is achieved by growing the SL on thick $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layers, deposited on GaAs substrates. By varying the thickness of the alloy well layers, we demonstrate the predicted crossover of the lowest-energy emission from a direct, electron-to-heavy-hole transition to a spatially indirect (type-II) electron-to-light-hole transition. In addition, we use PLE spectroscopy, to characterize the strain relaxation in the $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layers. The compressive strain in the alloy buffer layer produces a splitting between the light- and heavy-hole exciton peaks observed in the PLE spectra, and hence we measure directly the residual strain as a function of $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ thickness. Even in layers that are several micrometers thick, we still see evidence for the presence of strain, suggesting that, at small strain energies, dislocations are an inefficient means of relieving strain on a macroscopic scale. Our PLE results are confirmed by x-ray-diffraction measurements on the same samples. The degree of residual strain determined by mapping the x-ray intensity is in excellent agreement with our PLE values. Residual compressive strain in the buffer layer produces significant modifications of the electronic properties of the overgrown SL layers, and must be taken properly into account in order to explain our experimental results.

I. INTRODUCTION

Optical measurements on (In,Ga)As-GaAs strained-layer superlattices (SL's), grown on GaAs substrates, provide a means of characterizing and understanding the electronic properties of one of the simplest III-V strained semiconductor SL systems. In such structures the alloy layers are under biaxial compression in the layer planes (xy) and are subjected to a shear, uniaxial stress in the growth direction (z), while the binary layers are unstrained. The net effects of the hydrostatic and uniaxial components of the strain are to increase the (In,Ga)As band gap and to remove the zone-center degeneracy of the light- and heavy-hole valence bands such that the heavy-hole band is at a higher energy than the light-hole band. This produces significant modifications of the electronic properties of quantum-well and SL structures. For example, it has been demonstrated that a mixed, type-I, type-II band alignment exists for the heavy and light holes,¹ respectively.

In this paper we present results of an optical investigation of (In,Ga)As-GaAs SL structures in which compressive and tensile strain are incorporated simultaneously in alternate layers. This is achieved by growing $\text{In}_x\text{Ga}_{1-x}\text{As-GaAs}$ SL's on thick $\text{In}_y\text{Ga}_{1-y}\text{As}$ buffer layers, deposited on GaAs substrates. For our samples, in which $x > y$, the strain configuration is such that the alloy SL layers are in biaxial compression, while the binary layers are under biaxial tension. The investigations have two facets. Firstly, we use photoluminescence excitation

spectroscopy to characterize the residual strain in the buffer layers as a function of their thickness, and secondly we study the effects of the alternating strain on the electronic properties of the SL structures.

Growth of a buffer layer of (In,Ga)As which is sufficiently thick as to be completely relaxed is not a trivial matter. Clearly the layer thickness must exceed a critical value L_c , to allow the strain to be relieved via the generation of misfit dislocations. The presence of dislocations can degrade the transport and optical properties of structures with layers which exceed L_c and many experiments which make use of this fact have been used to determine L_c for strained (In,Ga)As layers for a range of indium concentrations.²⁻⁷ As pointed out by a number of groups,^{6,7} there is a considerable discrepancy between values of L_c determined by different measurement techniques. Good agreement was found with Matthews and Blakeslee's theory⁸ for strained single layers measured by photoluminescence (PL) techniques.⁵ However, much larger values of L_c have been measured via techniques such as double-crystal x-ray diffraction (XRD). For example, Orders and Usher⁹ report values of L_c in good agreement with People and Bean's energy-balance model.¹⁰ These conflicting results can be explained by the varying degrees of sensitivity of the different measurement techniques to the presence of dislocations. A full discussion can be found in Ref. 6. The calculated critical layer thickness for an (In,Ga)As layer with an indium concentration of 5% varies from $\sim 0.04 \mu\text{m}$ (Ref. 8) to $\sim 1 \mu\text{m}$.¹⁰ Beyond the critical thickness there is no sharp

transition from the strained to the perfectly relaxed state, only gradual relief which occurs over a finite thickness and is dependent on the degree of strain in the system. The total thickness needed to produce material with the relaxed lattice parameter increases rapidly as the indium concentration, and hence the lattice mismatch decreases. Orders and Usher⁹ measure this transition in $\text{In}_y\text{Ga}_{1-y}\text{As}$ as 0.8, 0.6, and 0.3 μm for $y=0.07, 0.14,$ and 0.25, respectively. In this publication, we report measurements of the residual strain in $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ layers over a range of thickness from 0.5 to 6.0 μm .

Unlike the case of $\text{In}_x\text{Ga}_{1-x}\text{As-GaAs}$ SL's grown on GaAs where, as already discussed, only the alloy layers are strained, if the SL is grown on an $\text{In}_y\text{Ga}_{1-y}\text{As}$ buffer layer, then providing $x > y$, the GaAs layers are in biaxial tension and the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers are biaxially compressed. In this strain configuration, the highest valence band in the (In,Ga)As is the heavy hole while the light hole is highest in the GaAs layers. Figure 1 is a schematic illustration of the band alignment for 10% (In,Ga)As wells and GaAs barriers, deposited on "relaxed" 5% (In,Ga)As, assuming a band-offset ratio for the strained electron to heavy-hole gaps of 66:34. The choice of band-offset ratio is discussed in a later section. In this configuration the electrons and heavy holes are confined in the (In,Ga)As layers, producing a type-I arrangement, whereas the light holes are confined in the GaAs, making them type II with respect to the electrons. In these investigations we aim to exploit this strain configuration, by designing structures in which the hole-state energies are reversed and the lowest-energy emission is the spatially indirect, type-II electron-to-light-hole transition. There have been a number of other studies in different material systems which all use biaxial tensile strain in a quantum well to produce a reversal of the hole states. However, in previous illustrations of this

phenomenon the light hole always remains type I with respect to the electrons. In the GaAs-based materials, this includes optical investigations of GaAs-(InAlGa)As multiple-quantum-well structures¹¹ and of Ga(As,P)-(Al,Ga)As single quantum wells¹² grown on GaAs substrates. Similar results have also been demonstrated in (In,Ga)As-(In,Al)As quantum wells grown on InP substrates¹³ and in GaAs-(Al,Ga)As structures grown on Si substrates.¹⁴ In addition, Sauvage and co-workers¹⁵ have investigated (In,Al)As-GaAs SL's grown of buffers of (In,Al)As, deposited on GaAs substrates. In this last example, it is the lattice parameter of the (In,Al)As buffer, parallel to the interface plane, which is commensurate through the whole SL and which controls the degree of strain. We note that Sauvage and co-workers¹⁵ comment on the difficulty they found in obtaining complete plastic relaxation of the (In,Al)As buffer, even though the layer was $\sim 0.55 \mu\text{m}$ thick. This point is particularly pertinent to our investigations because although we are working in a different material system, the degree of lattice mismatch in the two cases is very comparable.

II. SAMPLE DETAILS AND EXPERIMENTAL TECHNIQUES

The structures used in these investigations were all grown by molecular-beam epitaxy in a Varian modular Gen II machine, at a substrate temperature of $\sim 530^\circ\text{C}$. The layers were deposited on undoped (001) GaAs substrates, rotated at ~ 20 rpm to maintain lateral uniformity over the wafer. The growth rates of GaAs and (In,Ga)As were measured using the reflection high-energy electron-diffraction (RHEED) technique on a GaAs monitor slice prior to the growth of the samples. Using this technique, the fluxes of indium and gallium were adjusted so that the nominal indium fraction in the SL alloy layers would be ~ 0.10 . For the growth of the (In,Ga)As buffer layers the fluxes are chosen to produce nominally half this indium fraction, ~ 0.05 . The samples used to study the strain relaxation in thick layers of (In,Ga)As consisted of either a 0.5, 1.0, 3.0, or 6.0 μm $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer, ten repeats of either 90- or 100- \AA $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells separated by 100- \AA GaAs barriers, and a 200- \AA GaAs capping layer. The second set of samples, which was grown primarily to study the electronic properties of SL structures grown on (In,Ga)As, followed a similar growth sequence. In this case all the SL's were grown on 1.0- μm $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layers. The GaAs barrier thickness was fixed at 100 \AA for samples with $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ well layers of 100, 90, 70, or 50 \AA , but was increased to 400 \AA for samples with 30- and 15- \AA wells.

Photoluminescence (PL) and photoluminescence excitation (PLE) spectra were recorded at 4 K. The PL measurements were made using an Ar^+ -pumped solid-state Ti:sapphire laser, set above the GaAs band gap, at 8100 \AA , as the excitation source. The same laser system provided the tunable source for the PLE measurements. In addition, we performed circularly polarized PL (PPL) and circularly polarized PLE (PPLE) measurements¹⁶ in which the linearly polarized laser light was chopped by an oscillating stress plate to produce alternating σ^+ and

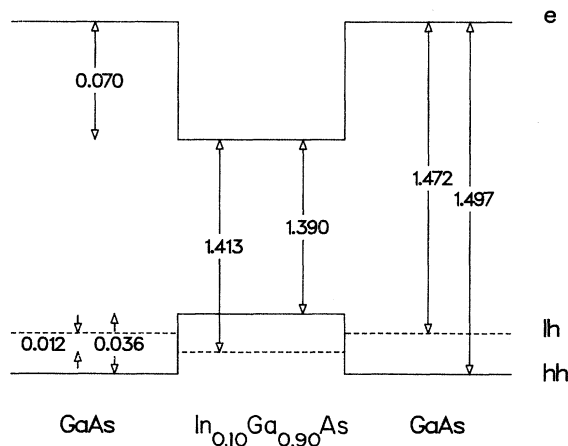


FIG. 1. Schematic band alignment of electrons (e), heavy-holes (hh), and light-holes (lh), for $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and GaAs barriers if deposited on relaxed $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$, assuming a band-offset ratio for the strained electron to heavy-hole gaps of 66:34. The calculated band gaps and barrier heights are in electron volts.

σ^- excitation. In these measurements we selectively detect changes in one sense of the circularly polarized emission, with the phase of the system arranged so that transitions involving heavy holes produce peaks in the spectrum, while light-hole transitions appear as dips, thus providing valuable information to aid with the assignment of the spectral features.

The structural characterization of these samples was carried out by mapping the scattered x-ray intensity, collected on a multiple-crystal multiple-reflection diffractometer,¹⁷ to produce a high-resolution image of reciprocal space.¹⁸ The intensity profiles close to the asymmetric 115 reflections were measured for each sample to obtain the lattice parameter parallel and perpendicular to the interface as well as the extent of the relaxation. Therefore the indium composition in the layer, and the depth profile of the relaxation, could be determined.¹⁷ The intensity distribution around the symmetric 004 reflection was also measured to confirm that there was no relative tilting between the layer and the substrate.

III. STRAIN RELAXATION IN (In,Ga)As BUFFER LAYERS

To characterize the strain in thick (In,Ga)As layers via optical techniques we have studied samples in which SL layers are grown on $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffers of varying thickness. The SL region of the structure acts as an efficient trap for the photoexcited carriers and in all the samples recombination in this region gave rise to a strong signal in the PL spectra, due to the ground-state electrons and heavy holes, $e1\text{-hh}1$. By monitoring changes in this luminescence intensity as the excitation energy is varied we measure a PLE spectrum of each sample. This reveals information about the excitonic transitions associated with the SL, and, because the structure is electronically coupled, also measures directly transitions associated with the buffer layer.

In Fig. 2 we show the PLE and PPLE data for a sample which consists of a SL with nominally 90-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 100-Å GaAs barriers, grown on a 1- μm $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layer. The PL spectrum of this sample reveals an emission line at ~ 1.399 eV with a measured linewidth of ~ 6 meV. The PLE spectrum, recorded at a detection energy of 1.395 eV, is shown as the upper curve in Fig. 2, while the polarized PLE data, recorded at the peak of the emission, appear immediately below. In the PLE spectrum we identify three features associated with excitonic transitions in the SL, labeled I, II, and III, and two peaks, α and β , which we believe originate from the buffer layer. Detailed investigations of the SL transitions have been made for a set of samples in which the well width is varied, but we reserve discussion of these results for the following section. Here we focus on the assignments of peaks α and β . These features appear in almost the same energy position in all the samples we have studied as part of these investigations. In all cases, α is reproduced as a peak in the PPLE data, while β appears as a clear dip in the polarized spectra. This allows us to identify α as a transition involving heavy holes

and β as a light-hole feature. In this sample the energy splitting between α and β is measured as 7.7 meV.

We reiterate at this point that to make inferences about the absorption peaks in the buffer layer we have recorded the PPLE signal by detecting at the $e1\text{-hh}1$ transition in the (In,Ga)As quantum wells. This means that in addition to the three usual steps in the PLE process, namely, creation relaxation and recombination, a further intermediate step is involved—the tunneling of at least one of the carriers through the 100-Å barriers. In the simplest model one might expect this carrier to be the electron and indeed the observation of a polarized PLE signal is consistent with this supposition. In the most common picture of the observation of a polarization-dependent spectrum it is assumed that the excited hole population loses its spin memory completely while the electron spin is retained during the relaxation and recombination steps.¹⁹ This common assumption explains most, but not all,²⁰ of the features seen in the PPLE spectra of quantum wells and is sufficient to qualitatively explain our observations providing we further assume that the electron population retains its spin memory during the non-resonant tunneling process.

To be certain the transitions α and β are associated with the buffer layer, we have grown a number of samples without SL overlayers. These structures consist simply of thick $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ layers, deposited on GaAs substrates. The luminescence efficiency of these structures is at least two orders of magnitude lower than from the SL samples; nevertheless, PLE and PPLE measurements on a 1- μm layer revealed the same features, α and β , having heavy- and light-hole character, respectively. Their ener-

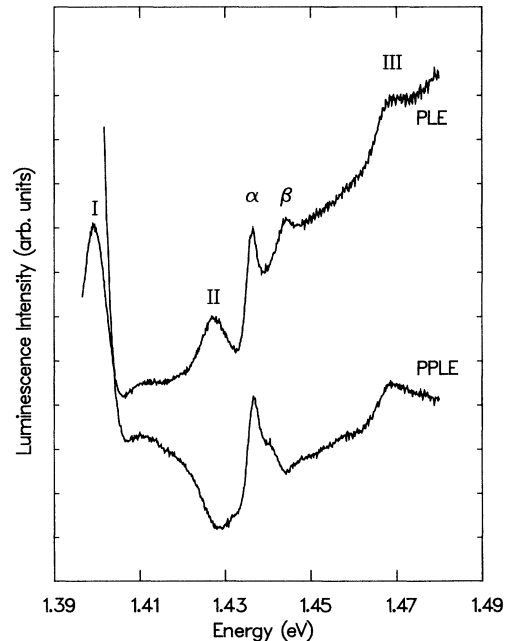


FIG. 2. Photoluminescence excitation (PLE) and circularly polarized PLE (PPLE) spectra, recorded at 4 K, from the superlattice with nominally 90-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 100-Å GaAs barriers, deposited on a 1- μm $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layer.

gy positions in this sample are within 2 meV of those observed in Fig. 2. Therefore we can, without any ambiguity, assign peaks α and β to the heavy-hole and light-hole exciton transitions in the bulk $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ layer. The observation of this pair of transitions, brought about by the lifting of the valence-band degeneracy, implies that the buffer layer in this sample is not perfectly relaxed, but instead must incorporate a degree of residual compressive biaxial strain and uniaxial extension.

The identification of the heavy- and light-hole buffer transitions provides two important pieces of quantitative information. The midpoint between peaks α and β measures, for small strains, the position of the hydrostatically strained $\text{In}_y\text{Ga}_{1-y}\text{As}$ band gap, and the splitting between the peaks measures the uniaxial extension, which lifts the degeneracy of the valence-band states. Combined, these two measurements determine uniquely values of the composition y and the lattice constant a_m of the partially relaxed buffer layer. (Details of the calculations including the values of deformation potentials and elastic constants used are given in Sec. IV.) Using the derived values of y and a_m we calculate the residual biaxial strain in the layer ϵ , which we define as

$$\epsilon = (a_y - a_m) / a_m,$$

where a_y is the in-plane lattice constant of “relaxed” $\text{In}_y\text{Ga}_{1-y}\text{As}$ on GaAs.

To explore further the extent to which $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ retains some degree of strain, even when the layer is far in excess of the theoretical estimation of the critical thickness, we have measured a set of four samples in which the thickness of the nominally 5% (In,Ga)As buffer is either 0.5, 1.0, 3.0, or 6.0 μm . For these investigations we again make use of SL overlayers as a means of measuring the buffer-layer transitions. In all four structures the SL is built from 100-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 100-Å GaAs barriers. The PLE spectra of all four samples are qualitatively similar to the results already illustrated in Fig. 2. Again, we identify both SL excitonic peaks and features associated with the buffer layer. For each sample we measure the splitting between the heavy- and light-hole buffer peaks, α and β , and the energy of

their midpoint. In all four structures we determine the indium fraction in the $\text{In}_y\text{Ga}_{1-y}\text{As}$ buffer layer to be $y \sim 0.05$. Using this composition and the derived values of a_m we calculate the residual strain in the buffer layers. We plot our results, as a function of the (In,Ga)As buffer thickness, in Fig. 3. The data point at 0 μm is a calculation of the biaxial strain incorporated in an $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ layer deposited on GaAs, assuming the alloy is perfectly elastically strained, i.e., $a_m = a_{\text{GaAs}}$. The line drawn through the data points is only a guide to the eye. From this data it is immediately apparent that in all the samples studied, we have exceeded the theoretical estimation of the critical thickness for 5% (In,Ga)As. Over the first 0.5 μm the strain has fallen by $\sim 65\%$. However, as the buffer-layer thickness is increased there is only a very gradual decrease in the residual strain, and even in layers which are 6.0 μm thick we still see evidence for the presence of strain. This implies that while dislocations must be present in the material, and locally they act to relax the lattice, on a macroscopic scale there must still be large regions of the crystal which remain under biaxial compression. Therefore, in these samples it appears that for small strain energies, i.e., when the residual strain in the layer falls below about 0.10%, dislocation generation is a very inefficient means of facilitating stress relief.

X-ray-diffraction (XRD) techniques have also been used to characterize the residual strain in these layers, although these measurements are only used as a means of confirming the optical data. The XRD maps²¹ also show that the (In,Ga)As buffer layers are only partially relaxed, even at 6- μm thickness. The in-plane lattice parameter for each sample was determined from the difference between the diffraction peaks of the buffer layer and the substrate parallel to the surface. The residual strain at 0.5, 3, and 6 μm amounts to 0.142%, 0.111%, and 0.044%, in good agreement with our PLE results in Fig. 3. Contrary to the XRD measurements of Orders and Usher,⁹ who suggested that (In,Ga)As layers with a nominal indium fraction of 0.07 are completely relaxed at thicknesses above 0.8 μm , our measurements show that even at 6 μm the average lattice constant of a 5% layer is only about 87% relaxed. By combining the measured lattice parameters perpendicular and parallel to the surface, and the appropriate elastic constants, we determine the lattice parameter for an equivalent free-standing buffer layer of (In,Ga)As, and hence determine the indium fraction in the layers. Since no significant variation in the perpendicular lattice parameter with depth could be measured when compared with the simulated diffraction profiles, it appears that the tetragonal distortion is uniform throughout the buffer layer. Furthermore, no difference could be measured between the in-plane lattice parameters for the superlattice and the buffer, indicating that the atomic planes are commensurate across this interface. A more detailed account of the x-ray scattering results will be published separately.²¹

Clearly, any degree of residual strain in the (In,Ga)As buffer layer will dramatically affect the electronic properties of the overgrown SL structure. In the ideal situation, in which the buffer is perfectly relaxed, and has an indium composition of 5%, then for a SL structure with ex-

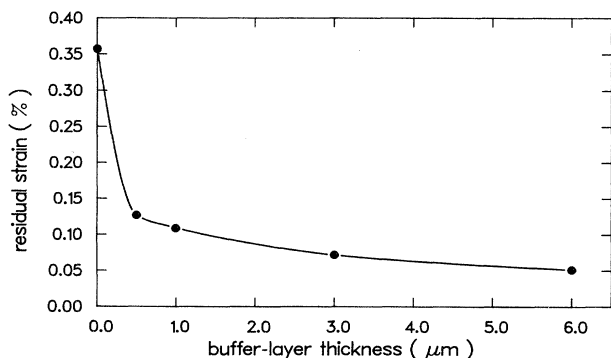


FIG. 3. Photoluminescence excitation measurements of the residual biaxial strain in $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ as a function of layer thickness. The solid curve is a guide to the eye only.

actly 10% indium in the well layers, separated by GaAs barriers, the resulting strain would be equal but opposite in alternate layers. The SL alloy layers would be under a compressive strain of 0.36% and the GaAs layers would be under a tensile strain also of 0.36%. However, if we take, for example, the case of 1- μm buffer, we measure a residual compressive strain in the layer of $\sim 0.12\%$. This leads to an increase in the compressive strain exerted on the (In,Ga)As SL layers to $\sim 0.48\%$ and a decrease in the tensile strain in the GaAs layers to $\sim 0.24\%$. These changes produce significant modifications of the electronic properties of the SL layers and must be properly taken into account in order to explain our experimental results.

IV. SIMULTANEOUS COMPRESSIVE AND TENSILE STRAIN IN SUPERLATTICES

In our investigations of the electronic properties of $\text{In}_x\text{Ga}_{1-x}\text{As-GaAs}$ SL's grown on thick $\text{In}_y\text{Ga}_{1-y}\text{As}$ layers we have studied a set of six samples, all grown on 1.0- μm $\text{In}_y\text{Ga}_{1-y}\text{As}$ buffers, with a nominal indium fraction of $y=0.05$. The width of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ wells is varied between samples from 100 to 15 \AA , with a fixed nominal indium fraction in the well layers of $x=0.10$. The PLE data on four of the samples are illustrated in Fig. 4. The well width of each sample is given in the figure and nominally these structures all have the same GaAs barrier width of 100 \AA . Included in this figure is the PLE spectrum shown in Fig. 2 for the 90- \AA sample. Omitted from the figure are the PPLE spectra, which have also been recorded for each of the samples. In all cases the circularly polarized experiment revealed similar

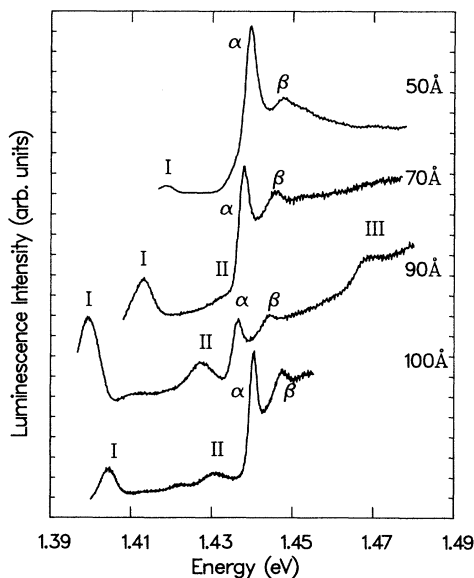


FIG. 4. Photoluminescence excitation spectra of superlattices deposited on 1 μm $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layers. The thickness of the $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells in each sample is indicated in the figure; all the samples have 100- \AA GaAs barriers.

data to that illustrated in Fig. 1. Peaks α and β , the heavy- and light-hole buffer transitions, are identified in each spectrum. The slight shift in energy position of these peaks can be accounted for by allowing small variations in the indium fraction of the buffer layers, between samples. To make a comparison with calculations of band-to-band transitions we assume an exciton binding energy for bulk $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ of 3.5 meV. From the energy positions of peaks α and β we determine the composition of the buffer layers to be $4.9 \pm 0.15\%$, for all the samples. The energy splitting between α and β varies between 6.8 and 7.8 meV for these samples, corresponding to small changes in the average in-plane lattice parameter of the buffer layers. From these measurements we calculate a residual compressive strain in the (In,Ga)As buffers of $0.11 \pm 0.01\%$ for the four samples.

We now turn our attention to the SL exciton transitions, labeled as I, II, and III. We compare the measured exciton positions with our envelope-function calculations. The effects of the uniaxial and hydrostatic components of the strain on the band gap are included in the calculation. We adopt the value of the GaAs deformation potential ($a = -8.6$ eV) used by Gershoni and co-workers to fit optical data on both (In,Ga)As-InP (Ref. 22) and (In,Ga)As-GaAs (Ref. 23) quantum-well structures. All other details of the calculation can be found elsewhere.^{24,25} To make a comparison between the exciton positions measured experimentally and our calculated band-to-band transitions we assume a fixed value of the binding energy for all the exciton states of 6 meV. This is in line with experimentally determined values of the $e1\text{-hh}1$ binding energy²⁶⁻²⁸ in (In,Ga)As-GaAs SL's grown on GaAs substrates. In order to fit the positions of the exciton transitions we have identified in the PLE data, we allow small deviations away from the nominal composition of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ SL layers. As already discussed, the composition and the residual strain of the buffer layer are measured directly for each sample. In some samples we also measure the tensile strain in the GaAs SL layers, but in all other cases we assume that the measured lattice parameter of the (In,Ga)As buffer, parallel to the interface plane, propagates through the whole SL. This assumption is supported by the XRD analysis of these samples which, as already discussed, indicates that indeed the SL overlayers are commensurate with the buffer material.

The best fit to all the experimental data is obtained using a band-offset ratio for strained $\text{In}_{0.10}\text{Ga}_{0.90}\text{As-GaAs}$ on $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$, electron to heavy-hole gaps, of 66:34. The choice of the band-offset parameter has been a matter of some controversy in the (In,Ga)As-GaAs system,²⁹ where the SL layers are deposited directly onto GaAs substrates. However, we,³⁰ and others,^{1,31} have found that in this system a band-offset ratio of $\sim 67:33$ has consistently produced an excellent fit to optical measurements of intersubband transitions. However, to date no measurements of Q_c have been reported for (In,Ga)As-GaAs SL's grown on (In,Ga)As. Qualitatively, it is not unreasonable to expect the value of Q_c in this strained configuration, which incorporates only a relatively small fraction of indium in the buffer layer, to be similar to the value determined for a SL deposited on

GaAs. Furthermore, this choice of Q_c is in good agreement with Van de Walle's model-solid theory³² of band lineups, from which Q_c is calculated to be ~ 0.64 for our material combination.

For all the data illustrated in Fig. 4, the lowest-energy transition has heavy-hole character and is almost coincident (within 1 meV) with the position of the dominant emission line measured in the PL spectra. We ascribe peak I to the ground-state electron to heavy-hole exciton transition, $e1\text{-hh}1$. A clear dip in the PPLE data of these samples (an example of which is shown in Fig. 1), appears at the position of peak II, allowing us to identify this transition as the electron to light-hole exciton, $e1\text{-lh}1$. A further prominent peak appears in the spectrum of the 90-Å sample, above the $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ band edge, labeled as peak III. This transition is reproduced as a peak in the PPLE spectrum, hence we associate this feature with an exciton transition having heavy-hole character. Comparison with our calculation leads us to assign peak III to the allowed $ec\text{-hh}1$ exciton state, where the notation ec implies that this is a transition involving the electron continuum states. An extensive investigation of features associated with transitions from confined states to continuum states in (In,Ga)As-GaAs structures (grown on GaAs) is described in Ref. 33. We do not expect to observe this feature within the spectral range we have measured in any of the other samples. Similarly, it is not surprising that the $e1\text{-lh}1$ transition is not clearly observed in the spectrum of the 50-Å sample. For a sample with these dimensions the light-hole peak is predicted to be in the same spectral region as the heavy-hole buffer transition, which clearly dominates the spectrum at these energies. However, we note a broadening on the low-energy side of peak α , which may represent a contribution from the light-hole exciton transition in this sample.

If the nominal structures of these samples were reproduced exactly, we would of course expect to observe a progressive shift of the $e1\text{-hh}1$ exciton peaks, shown in Fig. 4, to lower energy as the well width is increased. In practice this is not the case, and in order to explain the measured exciton positions we have to introduce small deviations from either the nominal layer thickness or the well composition. We obtain excellent fits to all the PLE data on these samples by using the measured compositions and in-plane lattice constants of the buffer layers, assuming the nominal well and barrier widths, and allowing a variation in the indium fraction in the wells of 0.12 ± 0.01 over the four samples.

As the thickness of the (In,Ga)As wells is progressively decreased the confinement energy of the electron and hole subbands increases until for samples with wells narrower than ~ 40 Å, the $e1$ energy level is calculated at an energy greater than the $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ conduction-band edge. Consider a SL sample with 30-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 100-Å GaAs barriers. For such a structure we calculate a miniband width for the first electron level of ~ 19 meV. This degree of electronic coupling between the layers means that if electrons are excited in the sample they will escape from the SL region of the structure and recombine via the lower-energy path, in the buffer layer.^{34,35} To enable us to monitor the positions of the

SL minibands in samples with thin (In,Ga)As wells we have designed structures in which the GaAs barrier thickness is increased to 400 Å. This reduces the coupling between the wells and allows us to measure luminescence from energy states above the buffer band

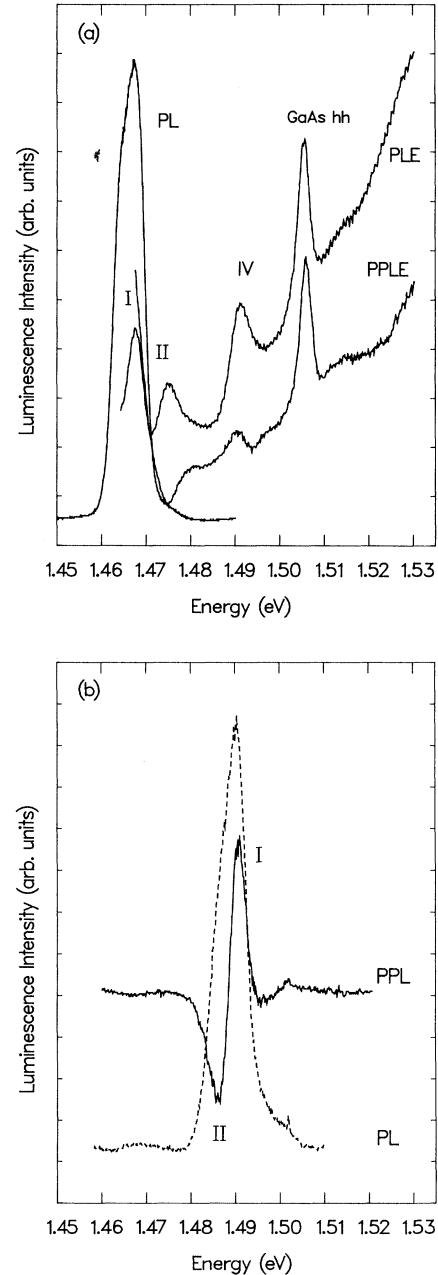


FIG. 5. (a) Photoluminescence (PL), photoluminescence excitation (PLE), and circularly polarized PLE (PPLE) spectra, recorded at 4 K, for the superlattice with nominally 30-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 400-Å GaAs barriers. (b) Photoluminescence (PL) and circularly polarized PL (PPL) spectra, recorded at 4 K, for the superlattice with nominally 15-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 400-Å GaAs barriers. Both superlattices were deposited on 1- μm $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ buffer layers.

gap.

In Fig. 5(a) we show the PL, PLE, and PPLE data on a sample with nominally 30-Å $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ wells and 400-Å GaAs barriers. The PL spectrum of this sample shows an emission line at ~ 1.468 eV with a measured linewidth of 7.3 meV. The slight asymmetry of the emission peak suggests that there is probably some small inhomogeneity in the sample, of either the composition and/or layer thickness, giving rise to more than one contribution to the exciton peak. The PLE spectrum, recorded with the detection set at 1.4617 eV, is shown as the upper curve in Fig. 5(a), while the polarized PLE data, which were recorded at the peak of the emission, appear immediately below. Again, the lowest-energy emission in this sample is the heavy-hole exciton, $e1\text{-hh}1$. The light-hole exciton, $e1\text{-lh}1$, can be identified from the dip in the PPLE spectrum at ~ 1.476 eV and is labeled in the figure as peak II. The splitting between the heavy- and light-hole states in this sample is measured to be only 8 meV. The shape of the PPLE spectrum near peak IV reveals that this feature is made up of two contributions; a heavy-hole transition at ~ 1.491 eV and a transition with light-hole character at ~ 1.494 eV. We consider firstly the assignment of the transition at ~ 1.491 eV. We ascribe this peak to the parity forbidden $e1\text{-hh}2$ exciton transition, which we have identified previously in (In,Ga)As-GaAs SL samples.³⁰ There are a number of possible reasons which may account for the appearance of this transition, including extraneous electric fields or grading of the indium composition, both of which may cause some asymmetry in the potential profile. Alternatively, deviations from the exact periodicity of the SL, or in-plane mixing of the light and heavy holes (in this case lh1 and hh2), can also be responsible for the breakdown of the selection rules.

This leaves the light-hole transition at ~ 1.494 eV and the sharp peak at ~ 1.505 eV in Fig. 5(a) still to be assigned. In these samples the GaAs SL layers are under biaxial tension so that the lattice parameter is extended in the layer plane. In this strain configuration the degeneracy of the valence-band states in the bulk material is lifted, resulting in a splitting of the light- and heavy-hole states and a decrease in the GaAs band gap. Therefore, in these samples we expect to see absorption due to both GaAs hole states in the PLE spectrum. We ascribe the transition at ~ 1.494 eV to the GaAs light-hole exciton, and the feature at ~ 1.505 eV to the GaAs heavy-hole exciton state. A measurement of the position and splitting of these exciton transitions provides an excellent means of determining the degree of strain incorporated in the GaAs layers. For this sample, the energy shift of the heavy-hole exciton from its unstrained position of ~ 1.515 eV (allowing an exciton binding energy of 4.2 meV), can be accounted for by a tensile strain in the GaAs layers of $\sim 0.16\%$. This produces a calculated splitting between the GaAs valence-band states of 10.6 meV, in excellent agreement with the splitting measured (~ 11 meV) in the experiment. Therefore, only half the anticipated tensile strain has been incorporated into the GaAs layers, suggesting that partial strain relaxation is significant in the samples where we have increased the

GaAs barrier thickness to 400 Å, which is of the order of the critical thickness for the onset of dislocations, at this lattice mismatch.⁸ Assuming that the measured GaAs lattice constant controls the strain in the whole SL structure, we obtain agreement, to within 2 meV, between the measured exciton positions, including peak IV, the $e1\text{-hh}2$ transition, and our envelope-function calculations by adjusting the indium composition in the well layers of this sample to be 11.5%.

The final SL sample in these investigations has nominally 15-Å (In,Ga)As wells with a nominal indium fraction of 0.10. Thick GaAs barriers (400 Å) are again used to limit the degree of electronic coupling between adjacent layers. The PL spectrum of this sample is illustrated in Fig. 5(b). It consists of an emission line at 1.4905 eV, with a measured linewidth of ~ 7.7 meV. The PLE spectrum of this sample shows only one additional feature, namely the heavy-hole exciton from the strained GaAs barrier layers, measured at ~ 1.504 eV. This corresponds to a decrease in the GaAs heavy-hole band gap of ~ 10.5 meV from the unstrained gap, brought about by a tensile strain in the GaAs SL layers for this sample of $\sim 0.18\%$. Far more interesting, however, is the circularly polarized photoluminescence (PPL) spectrum of this sample, also shown in Fig. 5(b). For all other samples, including the 30-Å quantum-well structure where some asymmetry of the PL spectrum was noted, PPL measurements simply reproduce the features observed in the unpolarized PL spectra, confirming the heavy-hole character of the lowest-energy transitions. However, the polarized PL data of the 15-Å quantum-well sample clearly resolve two strong features within the emission line, labeled as peaks I and II, which have different hole character. Peak II, the lowest-energy state in this sample, clearly has light-hole character, and is split from the higher-energy heavy-hole transition, peak I, by ~ 4.8 meV. We identify these transitions as the spatially indirect, type II, $e1\text{-lh}1$ exciton, and the direct, $e1\text{-hh}1$ exciton state, respectively, and are able to fit both their absolute energy positions, and the splitting, to within 0.5 meV using an indium composition in the well layers of 11.0% and the measured GaAs lattice parameter which controls the strain in the whole SL structure. From this experiment we can extract a measurement of the linewidth of the heavy- and light-hole excitons of less than or equal to 3.5 meV for the 15-Å quantum-well sample, compared to a linewidth of 7.3 meV for the 30-Å sample. This decrease in the PL linewidth with decreasing well width, for narrow wells, is characteristic of the (In,Ga)As-GaAs system and has been reported elsewhere.³⁶

From these measurements we estimate reversal of the hole states in these samples to occur at an (In,Ga)As thickness of ~ 20 Å. This is to be compared with a predicted crossover of ~ 48 -Å (In,Ga)As if the nominal strained GaAs barrier layers, are assumed. Clearly, this represents a significant modification of the electronic properties of the SL structures induced by the presence of a degree of residual strain and it must be taken into account in order to explain the experimental results properly.

V. SUMMARY

Using photoluminescence excitation spectroscopy we have studied the electronic properties of $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ - GaAs superlattice structures grown on thick $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ layers, deposited on GaAs substrates. The strain configuration in these structures is such that the $\text{In}_{0.10}\text{Ga}_{0.90}\text{As}$ layers are under biaxial compression, while the GaAs layers are in biaxial tension. By varying the thickness of the SL alloy layers, we have demonstrated the predicted crossover of the lowest-energy emission from a heavy-hole to a light-hole transition. This occurs at a well width of ~ 20 Å in these samples. Comparing the measured exciton positions and crossover point with our envelope-function calculations, we find the best fit to all the data is obtained using a band-offset ratio of $\sim 66:34$ for this system.

Furthermore, we have used PLE spectroscopy to characterize the strain relaxation in the $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$

buffer layers. The compressive strain in the buffer produces a splitting of the light- and heavy-hole exciton states which we have observed in the PLE spectra. Hence, we have measured directly the residual strain in the buffer, as a function of the thickness of the layer. These results are in excellent agreement with the values of residual strain we determine via x-ray-diffraction mapping techniques on the same samples. Even in layers which are several micrometers thick we still see evidence for the presence of strain, suggesting that when the residual strain in the layer falls below about 0.10%, dislocations are an inefficient means of relieving strain on a macroscopic scale.

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*Permanent address: University of Twente, Department of Applied Physics, 7500 AE Enschede, The Netherlands.

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