

Evolution of the electronic states of coupled (In,Ga)As-GaAs quantum wells into superlattice minibands

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We have used photoluminescence excitation (PLE) spectroscopy to study the evolution of the electronic states, and associated optical transitions, as the number of wells is increased from 2 to 5 to 20 in the strained (In,Ga)As-GaAs quantum-well system. The indium fraction in the wells was nominally 0.12. The PLE spectra of the two-well and five-well samples show strong $\Delta n = 0$ exciton transitions and further features associated with transitions from confined states to continuum states. When a superlattice (SL) is formed, in the 20-well sample, we see momentum-conserving optical transitions at the mini-Brillouin-zone center and edge. We have also identified transitions associated with M_1 critical points in the SL band structure. One of these is a $\Delta n = 0$ exciton resonance below the saddle point while the other is a $\Delta n \neq 0$ exciton which has become allowed due to momentum mixing of the light- and heavy-hole bands in the plane perpendicular to the growth direction.

I. INTRODUCTION

Low-temperature photoluminescence excitation (PLE) spectroscopy has been extensively used to characterize the structure of isolated and coupled quantum wells in both the GaAs-AlAs system¹⁻⁴ and more recently in the strained-layer combination of (In,Ga)As-GaAs.⁵⁻⁷ In the study of superlattice (SL), structures much has been reported on the structural and electronic properties of very-short-period SL's in the GaAs-AlAs near-lattice-matched system⁸ and recently in the technologically challenging, highly strained InAs-GaAs⁹⁻¹¹ system. Very recent work^{12,13} on the optical properties of GaAs-(Al,Ga)As has also revealed additional exciton features associated with the saddle points (M_1 critical points) that exist in the one-dimensional superlattice band structure.

As far as we are aware, little work exists which catalogues the changes of the electronic structure of quantum wells into their SL counterparts as one alters, not the dimensions of the component parts, but the number of repeats of the well and barrier "building block." One exception is the work of Morris *et al.*¹⁴ who have characterized, both optically and structurally, (In,Ga)As-GaAs "superlattices," as function of the number of periods in the structure. These authors studied samples grown by low-pressure metalorganic chemical vapor deposition on (001)-oriented GaAs substrates; the nominal In fraction was close to 0.16 and the well-to-barrier ratio was approximately constant at $\sim (70 \text{ \AA}) / (70 \text{ \AA})$. The number of repeats of the well-barrier structure was varied from five through 10 to 20. The authors claimed that the structures with only five and ten repeats were of high structural integrity, while the 20-repeat sample showed evidence of strain relaxation, with supporting evidence coming from an x-ray study of the samples. Optical transitions

were identified in the low-temperature PLE spectra of the 10-period sample which corresponded to transitions both at the center of the one-dimensional (1D) mini-Brillouin-zone and at the zone edge, some of the transitions being forbidden by a simple symmetry argument. If one assumes that the band offset ratio between the strained electron to heavy-hole gaps of (In,Ga)As and GaAs is 67:33 at this indium fraction^{15,16} ($x \sim 0.16$) and calculates the predicted band widths for the $n = 1$ electron states, then one arrives at values of ~ 7.5 meV. Clearly, the nonzero bandwidth indicates that the system is a coupled one, and even for the five-repeat structure the separation between the hybridized, isolated well electron states of ~ 1.5 meV suggests that this structure will be quite a good approximation to a superlattice for these carriers.

In the work we present here we have designed the (In,Ga)As-GaAs samples so that the $n = 1$ electron minibandwidth for an infinite periodic structure would be ~ 20 meV. We have studied samples that have 2, 5, and 20 (In,Ga)As layers, all with a nominal indium fraction of ~ 0.12 . In contrast to other work, this particular set of samples gives us the opportunity to observe the hybridization of the individual, isolated quantum well eigenfunctions into the extended miniband states of the SL. Furthermore, we can follow the evolution of states above the barriers as continuum resonances are lost and forbidden gaps and unconfined minibands appear in the carrier dispersion.

II. GROWTH AND EXPERIMENTAL DETAILS

The coupled quantum well samples used in this study were grown by molecular-beam epitaxy in a Varian Associates Modular Gen-II system. A general description of the growth conditions used has been given in previous

publications.^{6,7} The particular samples studied here had the following nominal structure: (a) a 1- μm GaAs buffer, (b) either 2, 5, or 20 repeats of 25- \AA (In,Ga)As quantum wells, separated by 100- \AA GaAs barriers, and (c) a GaAs top capping layer of 0.24 μm , 0.21 μm or 200 \AA for the 2-, 5- and 20-repeat samples respectively. The substrate temperature during growth was nominally 580 C which is a temperature where significant indium reevaporation occurs.¹⁷ To compensate for this loss, an increased indium flux was used. Growth rates for both GaAs and (In,Ga)As were measured using the reflection high-energy electron diffraction (RHEED) oscillation technique on a GaAs monitor slice prior to the growth of the quantum-well samples. Using this technique, the incident fluxes of indium and gallium were adjusted so that the nominal In fraction in each sample ought to have been the same and have a value of ~ 0.12 . None of the layers was intentionally doped.

The PLE and circularly polarized PLE (PPLE) measurements were made using an Ar^+ -pumped dye (styryl-9) laser as the tunable excitation source. PPLE is a useful technique for distinguishing luminescence associated with either light or heavy holes. Details of the technique can be found in Ref. 1. In our PPLE experiments, the linearly polarized laser light was chopped by an oscillating stress plate to produce alternating σ^+ and σ^- excitation. The spectra were recorded at the $e1\text{-hh1}$ exciton position, selectively detecting only changes in one sense of the circularly polarized emission. We arranged the phase of the detection system so that this heavy-hole signal gave rise to a peak in the PPLE. Higher energy transitions involving heavy-hole states will therefore result in an increase in the PL intensity and hence a peak in the PPLE spectrum. Conversely, the participation of light-hole states will decrease the PL intensity, producing dips.

III. RESULTS AND DISCUSSION

The PLE spectra are presented in Figs. 1(a)–1(c) for the samples with 2, 5, and 20 wells, respectively. In all cases the detection energy was set in the low-energy tail of the $e1\text{-hh1}$ photoluminescence at (a) 1.4753 eV, (b) 1.4686 eV, and (c) 1.4704 eV. The circularly polarized photoluminescence excitation (PPLE) spectra for the samples are represented by the dashed lines in this figure.

A. Two-well sample

Even though we refer to this sample as having two wells, with our supposed strained electron to heavy-hole band offset ratio of 67:33, the sample is only a symmetric, double-quantum-well (DQW) system for the electrons and heavy holes. The electrons and light holes form a type-II configuration^{15,16} so that the light-hole valence-band edge is exactly out of phase with that of the heavy holes, making the (In,Ga)As layers barriers to light-hole motion in the growth direction (z direction). A schematic of the conduction- and valence-band-edge configurations for this sample is shown in Fig. 2(a). The structure is simultaneously a symmetric double-well system for electrons and heavy holes and a double-barrier system for the light holes.

Given this band-structure arrangement and our assumptions about the magnitude of the various potential steps, we can make use of some straightforward quantum mechanics¹⁸ to predict the allowed optical transitions. Taking transitions involving electrons and heavy holes first, we find that the eigenfunctions for electrons and heavy holes in this DQW structure are simply the symmetric and antisymmetric combinations of the eigenfunctions of the isolated QW's, the symmetric combination having the lower-energy eigenvalue. Allowed optical transitions should only occur between those energy levels

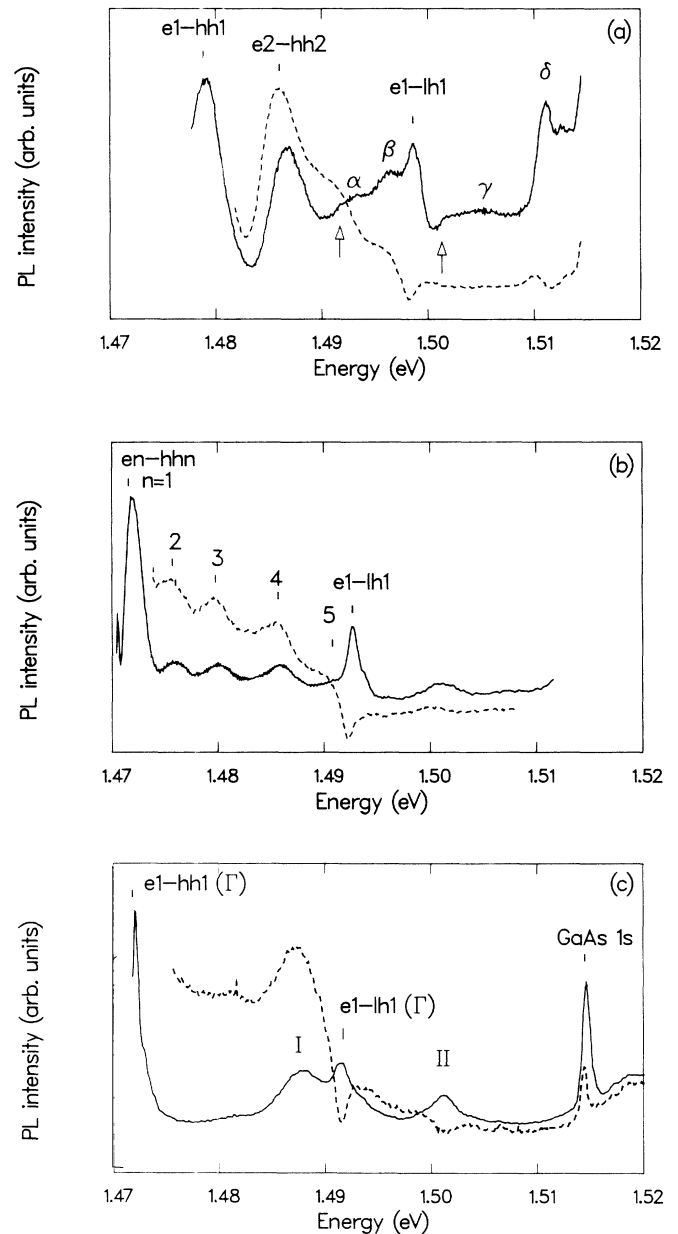


FIG. 1. Low-temperature (~ 4 K) photoluminescence excitation (PLE) spectrum (solid curve) and circularly polarized photoluminescence excitation (PPLE) spectrum (dashed curve) from (In,Ga)As-GaAs quantum well samples with (a) 2 (In,Ga)As layers, (b) 5 (In,Ga)As layers, and (c) 20 (In,Ga)As layers.

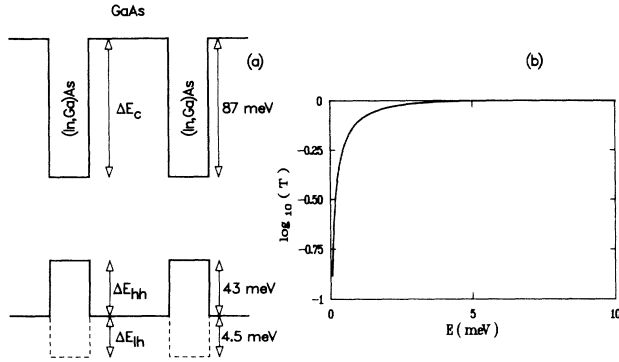


FIG. 2. (a) Schematic (not to scale) of the conduction- and valence-band edges for the two-well sample assuming a strained conduction-band offset fraction of 0.67 and (b) the energy spectrum for the probability of transmission of a light hole through the structure. For an indium fraction of 0.11 the light-hole barrier is ~ 4.5 meV in magnitude.

whose envelope functions have the same parity. In addition to the parity allowed transitions between confined states of the DQW, there also exists the possibility of allowed optical transitions between confined electron (heavy-hole) states and the heavy-hole (electron) continuum in the GaAs. The situation for optical transitions involving light holes is somewhat different. The (In,Ga)As layers are barriers for the light holes. With an offset ratio of 67:33 and using the deformation potentials suggested by Gershoni *et al.*^{5,19} we estimate that for an indium fraction of ~ 0.11 , the light-hole barriers are only ~ 4.5 meV high. We have calculated the transmission coefficient through the nominal $(25 \text{ \AA})/(100 \text{ \AA})/(25 \text{ \AA})$ double-barrier structure and find, as expected, no sharp peak in the transmission coefficient [see Fig. 2(b)]. The combined effects of a light-mass particle, low potential steps, and thin barriers smear out any sharp features, making the influence of the (In,Ga)As only the weakest of perturbations on the light-hole motion. It is reasonable to expect that the light holes in the system will behave in an almost three-dimensional (3D) manner, and we therefore expect to find optical transitions involving confined electrons and quasi-3D light holes in the GaAs.

Returning to Fig. 1(a), we identify the lowest-energy transition with the creation of the $e1$ -hh1 free exciton, where $e1$ and hh1 label the levels corresponding to the symmetric combinations of the isolated well eigenfunctions. The next peak is assigned to absorption between the higher-energy, antisymmetric heavy-hole and antisymmetric electron states of the DQW, which we call $e2$ -hh2. The PPLE spectrum for this sample allows us to confirm this assignment and to reveal the character of further transitions. The dip at 1.4986 eV indicates that this transition involves a light-hole state. This we assign to the $e1$ -lh1 free exciton which is a combination of the quasi-two-dimensional electron ground state with the quasi-three-dimensional light-hole state.

Further features, labeled by the lower-case Greek characters, are clearly visible in the spectrum. We have “fitted” the positions of the ($\Delta n = 0$) parity-allowed tran-

sitions that we have identified. We allow the layer thicknesses and indium fraction to vary between limits set by high-resolution x-ray diffraction on similar samples and the relatively small, but inevitable, errors associated with flux variations, etc., during growth at the substrate temperature of 580 C. Our “best fit” to this particular data is with an indium fraction of 0.105, a GaAs barrier of 110 \AA , and (In,Ga)As wells 25 \AA wide. To make the comparison we assumed a binding energy of ~ 8 meV for the $e1$ -hh1 exciton⁶ and a slightly smaller value for those transitions which we think involve extended or quasi-three-dimensional states, e.g., $e1$ -lh1. We assign peaks α and β to the exciton transitions $e1$ -hhc and ec -hh1, respectively. The notation hhc (ec) indicates that the state involved is that at (or very close to) the onset of the heavy-hole (electron) continuum states at the top of the heavy-hole (electron) barrier. The “continuum” states involved can be calculated by examining the transmission resonances in the energy range above the top of the barriers (see Fig. 3). These transmission resonances can give rise to sharp peaks that manifest themselves as exciton transitions in the PLE or absorption spectrum.^{18,20} Approximately 2 meV below the peak α , we see a distinct rise in the PLE signal. We speculate that this may be due to the excited states of the $e2$ -hh2 exciton and its attendant continuum. The rather broad feature labeled γ we think is probably associated with the transition $e2$ -hhc. On the low-energy side of γ (marked by an arrow), we again note a distinct rise in the PLE signal; given the quasi-3D nature of the $e1$ -lh1 exciton (and therefore its smaller binding energy), this feature could well originate from excited states of the $e1$ -lh1 exciton, unresolved from the continuum. The PPLE near the peak δ is a little ambiguous, but the increase at 1.51 eV suggests that the peak δ probably involves some heavy-hole contribution. The transmission resonances for the heavy-hole DQW

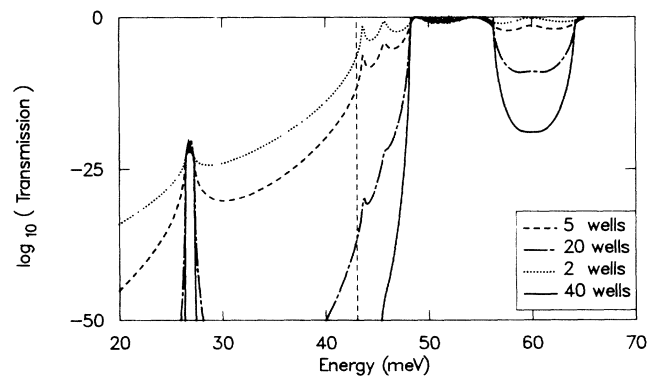


FIG. 3. Calculated transmission coefficient for the heavy holes through samples with 2, 5, 20, and 40 wells. Note that the resonances centered near 27 meV which are below the top of the barrier should reach unity; the fact that they do not is an artifact of the number of energy points used in the calculation. All other features are significant. The dashed vertical line at ~ 43 meV corresponds to the height of the heavy-hole potential step. The energy zero is at the position of the strained (In,Ga)As heavy-hole valence-band maximum.

system are shown in Fig. 3. We find a sharp resonance at the top of the heavy-hole well, a further slightly stronger resonance a couple of meV higher, and then almost unity transmission thereafter. The onset of this region of unity transmission in fact marks the energy at which a miniband will be formed in the SL which has the same well and barrier dimensions as the two-well sample. We therefore assign the peak δ to an exciton transition involving the state in the heavy-hole continuum at which unity transmission is reached and, most likely, from its energy position, the confined electron state $e2$.

B. Five-well sample

Figure 1(b) shows the PLE and PPLE spectra from the five-well sample. Again, strong $e1$ -hh1 and $e1$ -lh1 peaks are evident. Between these two sharp features there are three clear peaks, and we note an asymmetry of the light-hole peak on its low-energy side. Three peaks are also seen in the PPLE in this energy range, and a distinct change of slope can also be discerned prior to the light-hole dip. All these features are of heavy-hole nature, and we assign them to free excitons built from the optically allowed combinations of the confined excited states of the coupled five-well sample, i.e., en -hhn where n runs from 2 to 5. The calculated separation between the five individual electron levels ranges between 2 and 5 meV. This separation is too large to talk about this structure in terms of being a superlattice. Once more we have adjusted the nominal values of well and barrier width and indium fraction so that the envelope function calculations (including correction for exciton binding energies as in the two-well case) reasonably reproduce the strong exciton features. To describe this sample we used $x=0.11$, and (In,Ga)As width of 27 Å, and a GaAs barrier width of 110 Å. To complete the analysis of the spectrum, we need to interpret the peak at ~ 1.501 eV. In Fig. 3 we show the calculations of transmission probability for the heavy holes. If we compare the five-well (dashed) calculation to that of the two-well (dotted), then we find qualitatively that they are remarkably similar. Quantitatively, the strength of the first two resonances above the barrier is somewhat diminished. The behavior in the region between 57 and 65 meV indicates that part of the energy spectrum where a “forbidden” gap will appear in the eigenstates above the barrier as the number of periods increases. These calculations again indicate that there is a fairly high probability of finding hole states at or near to the top of the well whose wave functions will have some overlap with the confined electron states in the system. We therefore attribute the peak at ~ 1.501 eV to an exciton transition involving a confined electron state (most likely, $e5$) and the first resonant state in the heavy-hole continuum.

C. Twenty-well sample

The evolution of the system to a superlattice can be followed by further increasing the number of wells in the system. The final sample we have studied has a total of 20 wells (20 barriers for the light holes), and the PLE and PPLE spectra from this sample are shown in Fig. 1(c). Given the nominal well and barrier widths, we calculate that a SL with this period would have an $n=1$ electron

minibandwidth of ~ 20 meV, and that the top of the band would be at an energy smaller than the value of the 1D SL potential step ΔE_c . This means that the separation between individual electron states would be of the order of 1 meV—intuitively, a value small enough to talk about the electrons having formed a miniband rather than having to consider 20 individual eigenstates. The transition to a superlattice means that there now exists a 1D dispersion relation for the carriers in the growth direction. The wave vector in this direction, denoted by q , has now become a good quantum number. In a reduced zone scheme, forbidden gaps appear between bands alternately at $q=\pi/d$ and at $q=0$, i.e., at the 1D mini-Brillouin zone edge and zone center.

Further calculation supports the idea that the 20-well sample is a good approximation to a SL. In Fig. 3 we show the energy spectrum of the transmission coefficient for the heavy holes through the 20-well structure. A clear band of states appears (centered at ~ 27 meV) below the heavy-hole barrier (again 43 meV in magnitude). However, in contrast to the previous cases the transmission probability at the top of the well (near 43 meV) has dropped by ~ 25 orders of magnitude. The transmission probability only reaches unity 5 meV or so above ΔE_{hh} , and an allowed band of states extends up to ~ 57 meV. This is followed by a “forbidden” gap of ~ 8 meV in magnitude until the next band of allowed states is encountered. For illustrative purposes we have increased the number of wells to 40 and repeated the calculation. Qualitatively, there is no change in the spectrum from the 20-well calculation. Quantitatively, the only difference is the increase in the contrast between the allowed and forbidden regions of the spectrum, but, for example, there is no discernible change in the width of the forbidden gap between the allowed heavy-hole states. From the calculations presented above, showing the existence of forbidden gaps in the transmission through the 20-well sample, it is clear that the number of repeats is sufficient for the structure to be a good approximation to a superlattice.

In Fig. 1(c) the sharp peak at 1.4721 eV we assign to the recombination of free excitons at an energy smaller than the energy gap between the bottom of the $n=1$ electron miniband and the top of the $n=1$ heavy-hole miniband at the center of the 1D mini-Brillouin zone, i.e., $q=0$. We refer to this transition as $e1$ -hh1(Γ). At 1.4914 eV the PPLE spectrum shows a distinct dip—this transition we ascribe to the $e1$ -lh1(Γ) free-exciton transition. The further sharp peak at 1.5146 eV we assign to the 1s state of the bulk GaAs free exciton. Two further, somewhat broader, features appear at 1.4879 and 1.5011 eV which we refer to as peaks I and II, respectively.

The number of wells in the structure means we have to formulate an explanation of these features that properly takes into account the 1D periodicity of the structure and the evolution of the miniband dispersion. We can no longer talk about resonances in the continuum. The strongest direct optical transitions are expected at $q=0$ or at $q=\pi/d$. Furthermore, the parity of the hybridized, individual-well envelope functions in the z direction sug-

gests that strong transitions will only occur between subbands with the same index n . An intriguing feature of the SL band structure is the existence of M_1 critical points (saddle points) at the top (bottom) of the n th electron (hole) subband at $q = n\pi/d$. Zone folding back to the first 1D minizone means that the M_1 points alternate between $q = \pi/d$ and 0. The Coulomb interaction between electrons and holes associated with these saddle points can correlate their motion leading to the formation of excitonic resonances below the energy of the saddle point. In bulk materials Kane,²¹ for example, showed theoretically that these resonances manifest themselves as rather broad, asymmetric features. In Kane's treatment the resonance appears centered at an energy below the M_1 critical point which is almost identical to the energy at which the ground-state exciton appears below the minimum M_0 . Recently, saddle-point excitons in SL's have been studied theoretically²² and been seen experimentally in PLE studies of GaAs-(Al,Ga)As samples.^{12,13}

Peak I in the PLE spectrum of the 20-well sample is the exciton resonance associated with the M_1 critical points of the $n = 1$ electron and heavy-hole minibands at $q = \pi/d$. We denote this transition as $e1\text{-hh}1(\pi)$. The calculated sum of the $n = 1$ electron and heavy-hole miniband widths for this sample is ~ 16 meV, and we note that the separation between $e1\text{-hh}1(\Gamma)$ and $e1\text{-hh}1(\pi)$ is also ~ 16 meV. For this particular sample, therefore, the $e1\text{-hh}1(\pi)$ saddle point exciton resonance appears at an energy below the saddle point equal to the energy at which the $e1\text{-hh}1(\Gamma)$ exciton ground state appears bound below the fundamental gap. This is quantitatively what is expected from Kane's analysis and lends added weight to our assignment.

We think that the feature at 1.50 eV (peak II) again has its origins associated with an M_1 critical point. This time the saddle point involved is the M_1 critical point of the $n = 2$ heavy-hole miniband at $q = 0$, and we assign the peak to the parity forbidden $e1\text{-hh}2(\Gamma)$ exciton transition. This $\Delta n \neq 0$ transition may become allowed for a variety of reasons: (i) asymmetry in the potential profile caused by extraneous electric fields, grading of the indium profile, or local departures from a random alloy, (ii) some deviation from the exact periodicity of the SL or (iii) by analogy with the GaAs-(Al,Ga)As systems, in-plane mixing of the light and heavy holes.^{23,24} *A priori* we cannot

rule out any of these possibilities, and (i) and (ii) are distinctly possible given the state of knowledge about the molecular-beam-epitaxy growth of strained, indium-containing alloys, particularly at elevated growth temperatures. Point (iii) is the principal reason that $\Delta n \neq 0$ transitions are seen in the GaAs-AlAs system, and we suggest it is the most probable reason for the observation of this feature here. In addition, the lh1 and hh2 minibands cross in the SL direction, and it is likely that this degeneracy will cause strong mixing in the in-plane dispersion. The in-plane mixing of the light and heavy holes may also explain the somewhat ambiguous nature of the PPLE signal in the vicinity of the $e1\text{-hh}2(\Gamma)$ exciton. One further point remains to be made about this saddle-point feature. Unlike the $e1\text{-hh}1(\pi)$ exciton, which is a resonance, it is possible (because only the hole effective mass is negative) that the $e1\text{-hh}2(\Gamma)$ exciton will be a bound-state solution of the corresponding effective-mass equation that describes the exciton. A more systematic study of the physics of this particular transition is presented in another article.²⁵

IV. SUMMARY

By varying the number of repeats of the well and barrier building block of an (In,Ga)As-GaAs strained-layer structure, we have illustrated the evolution of the system from coupled quantum wells to a superlattice. The PLE spectra of the two- and five-well samples have shown strong $\Delta n = 0$ exciton transitions and further features associated with transitions from confined states to continuum states.

When a SL is formed in the 20-well sample, q becomes a good quantum number, and we see momentum-conserving optical transitions at $q = 0$ and π/d . We have also identified transitions associated with M_1 critical points in the SL band structure. One of these is a $\Delta n = 0$ exciton resonance below the saddle point, while the other is most likely a bound state of a $\Delta n \neq 0$ exciton, $e1\text{-hh}2(\Gamma)$, which has become allowed due to mixing of the light- and heavy-hole bands away from $k = 0$.

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