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Observation of luminescence from the 2*s* heavy-hole exciton in GaAs-(AlGa)As quantum-well structures at low temperature

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Photoluminescence experiments at low temperatures have revealed the existence of a previously unreported feature on the high-energy side of the $n=1$ (e-HH) 1s exciton recombination from GaAs-(AlGa)As multiple-quantum-well structures. Detailed circular-polarization and excitation-spectroscopy measurements have led to this feature being assigned to recombination of $n=1$ (e-HH) 2s excitons and free electron-hole pairs in the lowest subbands. In our samples, where there is no Stokes shift of the exciton lines, the energetic position of this peak relative to the main luminescence line gives a reliable measurement of the $n=1$ (e-HH) 1s exciton binding energy as a function of quantum-well width.

For many years now the optical properties of GaAs-(AlGa)As quantum-well structures have received extensive experimental and theoretical treatment. To realize a full understanding of these systems, fundamental and structural parameters such as the band offset, quantum-well width, exciton binding energies, and alloy composition must be determined. Two predominant optical techniques used to investigate quantum-well structures have been photoluminescence and photoluminescence excitation spectroscopy.[1] The dominant recombination mechanism in GaAs-(AlGa)As quantum wells at low temperatures (4 K) has been shown to be excitonic in character.[1] This process involves the recombination of $n=1$ electron-heavy-hole (e-HH) 1s excitons that are either free[1] or localized[2] at potential fluctuations due to variations in well width. This assignment is usually confirmed by the use of excitation spectroscopy, the exciton peak in luminescence being coincident with,[1] or red shifted from,[2] the peak position of the $n=1$ (e-HH) 1s exciton seen in the excitation spectrum. More recently, measurements[3,4] at low temperatures on GaAs-(AlGa)As quantum wells of thickness 200 and 120 Å have also revealed luminescence from the higher energy $n=1$ electron-light-hole (e-LH) excitons.

In this paper we report a previously unobserved peak in the low-temperature photoluminescence spectrum in GaAs-(AlGa)As multiple quantum-well structures with well widths L_z in the range 55–112 Å. We assign this emission to the overlapping contributions of excited states of the ($n=1$) e-HH exciton and free-carrier recombination. The assignment is confirmed both by comparison of the energetic position of the 2s exciton in the low-temperature excitation spectrum and by circular-polarization measurements. In samples where there is no Stokes shift of the exciton lines, its position relative to the main luminescence line gives a reliable measurement of the $n=1$ (e-HH) 1s exciton binding energy.

The multiple-quantum-well structures studied in this work were all grown in a Varian Gen II molecular-beam-epitaxy system. The layers were deposited on (001)-orientated semi-insulating GaAs substrates at a substrate temperature of 630 °C. The growth sequence was as follows: (a) 1.0 μm of GaAs buffer material, (b) 0.13 μm of (AlGa)As cladding, (c) 60 periods of GaAs wells and (AlGa)As barriers, and (d) 0.13 μm of (AlGa)As cladding. None of the layers was intentionally doped and the (AlGa)As barriers were thick enough to ensure the GaAs wells were electronically decoupled. The samples studied included a range of GaAs-Al$_x$Ga$_{1-x}$As multiple quantum-well structures with $x=0.35$ and well widths from 55 to 150 Å and a GaAs-AlAs multiple-quantum-well structure of 83 Å. The well width for each sample was determined by a detailed study of the low-temperature excitation spectra and by fitting the observed features to a theoretical model.[5] The quantum-well width and conduction-band offset are used as adjustable parameters.

The photoluminescence and excitation spectra were recorded with the samples mounted on the cold finger of a variable-temperature continuous-flow cryostat or in superfluid helium at 2 K. The samples were excited by an argon-ion-pumped tunable dye laser at power densities less than 5 W/cm^2 to avoid sample heating. In the photoluminescence experiments, the excitation energy of 1.771 eV was below the band gap of the cladding material. The luminescence from the front face was collected and analyzed by a double-grating Spex 1404 spectrometer and detected with a cooled GaAs photomultiplier and associated photon-counting electronics.

Photoluminescence circular polarization experiments have been shown[1] to be particularly useful in distinguishing different components in emission spectra associated with light or heavy holes. We performed these experiments by passing the linearly polarized laser light through an oscillating ($f=50$ kHz) stress plate to produce alternating σ^+ and σ^- excitation. Changes in one sense (either σ^+ or σ^-) of the circularly polarized emission were detected by passing the luminescence through a quarter-wave plate and linear polarizer and synchronously detecting changes in the polarization at 50 kHz.

The dominant feature in the low-temperature photoluminescence spectrum of these GaAs quantum wells is a line, of full width at half maximum (FWHM), in the range 1.6 meV at $L_z=150$ Å to 4.2 meV at $L_z=55$ Å, due to recombination of $n=1$ (e-HH) 1s excitons. In all our GaAs-(AlGa)As samples its position is coincident, within experimental error, with the spectral position of the $n=1$ (e-HH) 1*s* exciton in the excitation spectrum, and in the GaAs-AlAs sample it is within 1 meV. In addition, a further feature at higher energy is observed in the low-temperature spectrum of these structures. This is illustrated as a function of well width in Fig. 1. As previously observed[3,4] the high-energy line from the widest wells ($L_z=150$ Å) is due to $n=1$ (e-LH) exciton recombination. This assignment fits with the calculated HH-LH splitting and was confirmed by circular polarization experiments. However, as the well width is decreased the nature of the high-energy line changes. For the sample with 112 Å quantum wells, the high-energy feature has two contributions. Our circular polarization experiments, Fig. 2, show positive and negative changes in the sense of the circularly polarized emission ($\Delta\sigma^+$) of these two contributions, revealing that they are associated with recombination from heavy and light holes. Further decrease in the well width to 75 and 55 Å produces a single high-energy feature that is associated with recombination involving only heavy holes. Again this is demonstrated by the circular polarization experiment, Fig. 2, where a positive change in $\Delta\sigma^+$ is observed. Since the $n=1$ HH-LH splitting increases almost quadratically with decreasing well width, thermalization effects greatly reduce the emission associated with recombination of $n=1$ (e-LH) excitons which is too weak to be observed in the low-temperature spectra of the narrower samples.

The assignment of the main low-temperature emission line as recombination of $n=1$ (e-HH) 1*s* excitons has been confirmed by comparing its position with the exciton peak in the excitation spectrum. In the same way, information concerning the nature of the higher-energy emission lines observed in these samples is provided by comparing their spectral positions with detailed excitation spectra at the same temperature. This is illustrated in Fig. 3, where the comparison is made for an 83-Å GaAs-AlAs multiple-quantum-well sample. The high-energy luminescence peak lies within 1 meV of the feature in the excitation spectrum ascribed to the $n=1$ (e-HH) 2*s* exciton.[6] The small discrepancy is probably due to partial localization of the excitons at potential fluctuations due to steps at

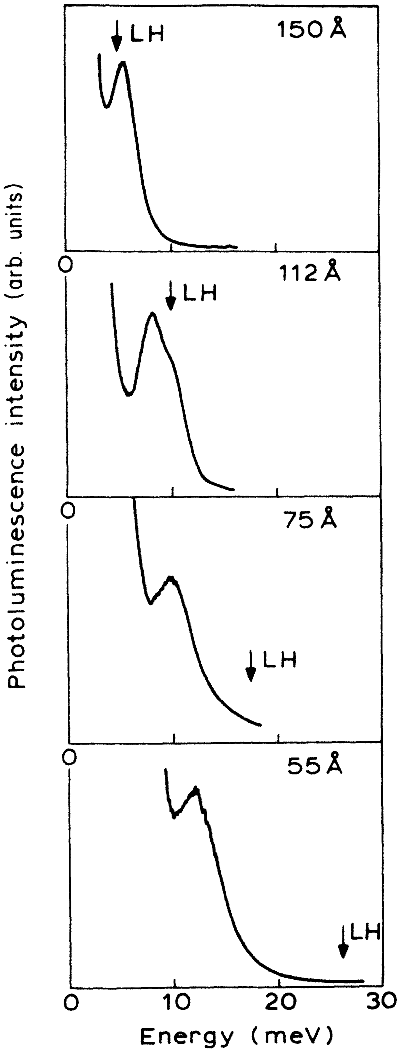

FIG. 1. Low-temperature (6 K) photoluminescence spectra for four GaAs-(Al$_x$Ga$_{1-x}$)As quantum wells with $x=0.35$. The zero of the energy scale is chosen to coincide with the peak of the $n=1$ (e-HH) 1*s* exciton line. The arrows indicate the position of the $n=1$ (e-LH) exciton peak, as measured in the corresponding excitation spectra.

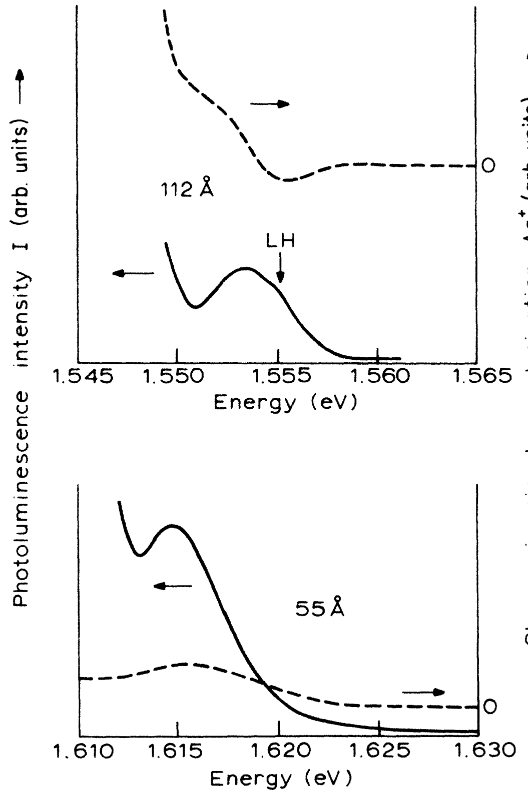

FIG. 2. Low-temperature (6 K) photoluminescence (solid line) and circular polarization (dashed line) spectra of the 112- and 55-Å quantum-well samples. Positive and negative changes in the sense of circular polarization correspond to heavy and light hole transitions, respectively.

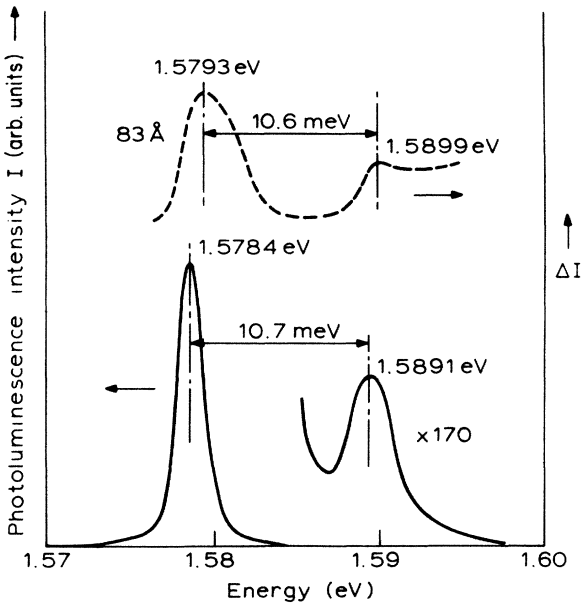

FIG. 3. Low-temperature (6 K) photoluminescence excitation (dashed line) and photoluminescence (solid line) spectra of the 83-Å GaAs-AlAs multiple quantum well. Note the clearly resolved $n = 1$ (e-HH) $2s$ exciton feature (Ref. 6) in the excitation spectrum and the small (< 1 meV) Stokes shift.

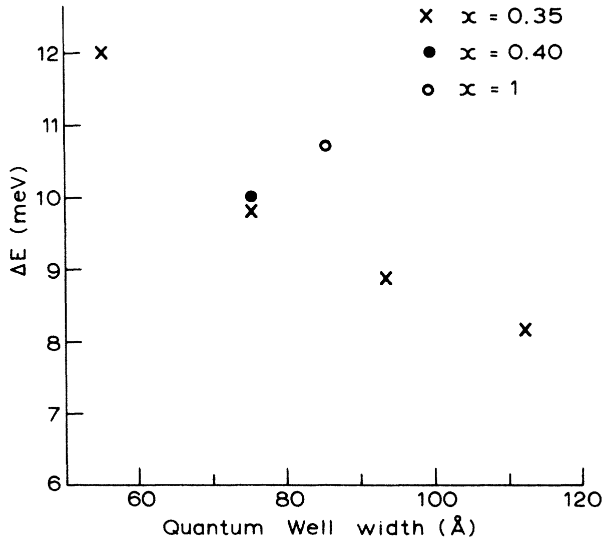

FIG. 4. Energy difference, ΔE of the $n = 1$ (e-HH) $2s$ exciton peak from the main $n = 1$ (e-HH) $1s$ exciton line as a function of well width.

the interface;[2] an identical shift is evident for the $n = 1$ (e-HH) $1s$ emission. Thus we propose that this new luminescence feature is associated with recombination of $n = 1$ (e-HH) $2s$ excitons.

However, we believe that this is not the only contribution to the observed emission. Unlike the shape of the $n = 1$ (e-HH) $1s$ line, the high-energy tails of these peaks are exponential, a characteristic of recombination involving at least one free carrier. This is most clearly observed in the 55 and 75 Å samples where there is no $n = 1$ (e-LH) exciton emission. Therefore, there is also a contribution to the emission from recombination of free electrons and holes in the lowest subbands. Assuming the free-carrier distribution obeys Boltzmann statistics, we can calculate an effective carrier temperature T_c of the system. For the 55-Å quantum well, under conditions of constant excitation energy (1.771 eV) we measure $T_c = 12$ K for power densities ≤ 5 W/cm^2. When the power density is increased, an increase in T_c to ~ 20 K at 120 W/cm^2 is observed, possibly reflecting the effect of lattice heating. The carrier temperature is also found to be a function of excitation energy and higher effective temperatures are measured if the excitation energy is increased. Clearly the system is not in equilibrium with the lattice.

Some understanding of these observations can be obtained by considering the carrier cooling mechanisms involved. It is well known[7] that in bulk GaAs polar optical-phonon scattering is an important energy relaxation mechanism, provided the excess energy for the electron and hole is higher than the threshold energy $\hbar\omega_0(\text{GaAs}) = 36.74$ meV. In the case where the excess energy is less than $\hbar\omega_0$, direct LO phonon emission is not possible, and then the dominant energy-loss mechanism is by acoustic phonon emission. In the quantum-well systems studied here the energy difference ΔE between the $2s$ emission peak and the $1s$ feature is between 8 and 12 meV, i.e., $\Delta E < \hbar\omega_0$. We believe that the slow cooling rate associated with acoustic-phonon emission accounts for both the relative strength of the observed high-energy line [which even at 2 K, is only 200–300 times weaker than the $n = 1$ (e-HH) $1s$ emission] and its associated effective carrier temperature.

The $n = 1$ (e-HH) $2s$ exciton feature observed in the excitation spectrum is only resolved from the continuum edge in our highest quality structures.[6] In emission, the excited state of the exciton is more routinely observed, in multiple-quantum-well samples of width 55-112 Å. In Fig. 4, we have plotted the energy difference ΔE between the high-energy luminescence and the $n = 1$ (e-HH) $1s$ exciton peak position as a function of quantum-well width. The position of the $n = 1$ (e-HH) $2s$ exciton peak can be measured to within an experimental error of ± 0.5 meV. Calculations of the $n = 1$ (e-HH) $2s$ exciton binding energy[6,8] predict a value in the range 1.5 ± 0.3 meV over the well widths studied in this work. Therefore, by adding this predicted $2s$ exciton binding energy to our measured values of ΔE, Fig. 4, we obtain a reliable measure of the $n = 1$ (e-HH) $1s$ exciton binding energy as a function of quantum-well width. As the well width decreases, for samples of Al fraction 0.35, ΔE increases smoothly, reflecting the increase of the $1s$ exciton binding energy with increased confinement energy. Also, we note the increase in ΔE when the barrier material is AlAs, reflecting the enhanced $1s$ binding energy when the barrier height is increased.

In conclusion, we have reported for the first time the observation of a luminescence line attributable to overlapping contributions from $n = 1$ (e-HH) $2s$ excitons and free carriers. This emission is observed routinely in our GaAs-(AlGa)As quantum wells with widths from 55 to 112 Å for a number of Al compositions. Combining the measured values of ΔE, the 1s-2s splitting observed in the photoluminescence spectrum, with predicted values of the 2s exciton binding energy[6,8] provides a simple and reliable method for measuring the $n=1$ (e-HH) 1s exciton binding energy.

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