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Well-resolved higher excited states of the light- and heavy-hole free excitons in a 225-Å $Al_xGa_{1-x}As$-GaAs multi-quantum-well structure

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Several excited states of the light- and heavy-hole free excitons have been observed in a 225-Å $Al_{0.35}Ga_{0.65}As$-GaAs multi-quantum-well (MQW) structure using photoluminescence excitation spectroscopy. The 2*s* and 3*s* states of both the heavy-hole free exciton, as well as of the light-hole free exciton, were identified. The measured values of the difference in energy between the ground state 1*s* and 2*s* state of both the heavy-hole and the light-hole free excitons agree with their calculated values. The binding energy of the 3*s* state is estimated using a hydrogen model, and the differences in energy between the 1*s* state and the 3*s* state of both the heavy-hole and the light-hole free excitons thus obtained are compared with their measured values. Magnetic field measurements were also made to verify the transition assignments.

The 2*s* excited states of the heavy- and light-hole free excitons have previously been observed in $Al_x Ga_{1-x}As$-GaAs quantum wells using photoluminescence excitation spectroscopy (PLE).[1-4] In two of these experiments, shoulders were observed on the high-energy side of the light- and heavy-hole free exciton transitions which were interpreted as representing the onset of the 2*s* excited states.[1,2] Another observation showed well resolved peaks on the high-energy side of the light- and heavy-hole free excitons which were identified as the excited 2*s* states of the light- and heavy-hole excitons.[3,4] Another experiment combining excitation spectroscopy with applied electric fields showed well resolved $n=2$ excited states for both the light- and heavy-hole free excitons.[5]

We have observed sharp-line structure associated with both the light- and the heavy-hole free excitons in multi-quantum-well (MQW) structures composed of Al_x-$Ga_{1-x}As$-GaAs in photoluminescence and reflection spectra.[6] These lines are very sharp (half widths as narrow as 0.02 meV in some samples) enabling one to observe excited states associated with each of the fine-structure components. We have observed the 2*s* and 3*s* excited states of the heavy-hole free exciton and the 2*s* and 3*s* excited states of the light-hole exciton in a 225-Å MQW. These states were observed using PLE techniques with the spectrometer positioned on one of the fine-structure components of the heavy-hole free exciton.

A number of calculations of the exciton binding energies in quantum wells have been performed in the last few years.[1,3,7-18] The energy separation that we measure between the 1*s* and 2*s* states agrees well with these calculated values. We are not aware of any measured or calculated values of the energy separation between the 1*s* and 3*s* states. We have made estimates of these differences and find them to agree with the measured values.

The different components of the sharp-line fine structure in the heavy-hole free exciton were examined by PLE. The different peaks in the fine structure arise from submonolayer changes in well size which are caused by the interface structure, as predicted.[19] The energy separations between the ground state and the excited states were essentially the same for all of the fine-structure components. This confirms that the exciton bonding energy does not change significantly from component to component.

The MQW structure used in this experiment was grown by molecular beam epitaxy (MBE) and had 30 cycles of 225-Å wells with 120-Å barriers. The Al concentration in the barrier material was 0.35. The structure was grown at 600 °C with a 2.8-μm-thick GaAs buffer. The MQW was excited with a tunable dye laser using styryl 9 dye which was pumped by an argon ion laser. The sample was mounted strain free on a sample holder which was immersed in liquid He and maintained at a temperature below 2 K by pumping on the liquid. A high-resolution 4-m grating spectrometer was used to monitor the degree of excitation of the various fine-structure components of the heavy-hole free-exciton emission spectra as the dye laser was scanned to higher energy.

A 225-Å MQW is a convenient structure to use for this experiment since the excited states of both the light- and heavy-hole excitons are on the high-energy side of the light-hole free-exciton ground state. Consequently, an advantage of this dimension well is that any interference from the light-hole free excitons is excluded.

The heavy-hole free-exciton ($E1$-HH1) photoluminescence spectra for this sample consisted of 7 sharp lines covering the spectral region from 8143.0 Å (1.52218 eV)

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to 8145.8 Å (1.521 66 eV). We now report the results obtained when PLE was used to investigate the excited states associated with both the light- and heavy-hole free excitons. The spectrometer was positioned on the highest-energy heavy-hole exciton sharp-line component (labeled 1 in the inset of Fig. 1) and the tunable dye laser was scanned over the excited-states region of both the light- and heavy-hole free excitons. The line at 1.5252 eV is the highest energy sharp-line component of the light-hole free-exciton ($E1$-LH1) structure as seen in photoluminescence. To obtain the remaining spectra (labeled 2–7 of Fig. 1), the spectrometer was positioned to the corresponding heavy-hole sharp-line component (given in the inset) and the region of the light-hole free exciton was rescanned by sweeping the dye laser. The heavy-hole exciton excited states are shown in Fig. 2. This spectrum was obtained by extending the energy scan of the dye laser past the light-hole feature for the highest energy heavy-hole fine-structure component (1 of Fig. 1). The transition observed at 1.5273 eV is suggested to be due to an exciton associated with the first conduction subband and second heavy-hole subband ($E1$-HH2). The $n=2$ excited state of the heavy-hole free exciton occurs at 1.5282 eV. The calculated value of the binding energy of the ground state of the heavy-hole exciton in this quantum well is 6.4 meV.[10] The calculated binding energy of the 2s state for this size well is 1.15 meV.[10] Thus, the separation of the 2s heavy-hole excited state from the 1s ground state is obtained by subtracting the 2s state binding energy from the 1s heavy-hole exciton binding energy which gives a value of 5.25 meV. From Fig. 2 the measured 2s state is 6.0 meV higher in energy than the 1s state, which is in quite good agreement with the calculated value. There are no calculated or measured values of the binding energy of the 3s excited state. Experimentally, the 3s state is 6.5 meV higher in energy than the 1s state. One can estimate the binding energy of the 3s state by assuming that it is $\frac{4}{9}$ of the binding energy of the 2s state (1.15 meV). The difference between the binding energies of the 1s and 3s states is then 5.9 meV which is in good agreement with the measured value.

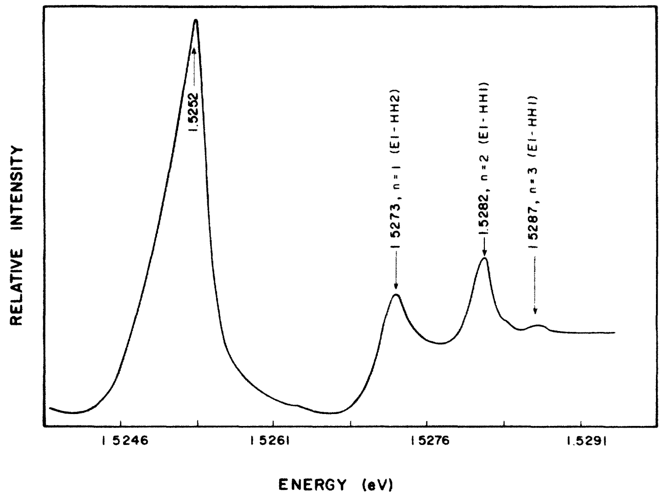

FIG. 2. The excited states of the heavy-hole exciton ($E1$-HH1) for a 225-Å MQW. This PLE spectrum is produced by measuring the relative excitation of the highest-energy component of the heavy-hole free exciton. The feature at 1.5252 eV is the highest energy component of the light-hole free exciton.

To gain additional evidence for the identification of the transitions shown in Fig. 2, the transitions were examined in a magnetic field. In this case, the magnetic field was applied parallel to the quantum-well layers and was varied from zero to 36 kG.[20] The lowest energy transition labeled $E1$-HH2 is a dipole forbidden exciton transition which is observed because of mixing of the first light- and the second heavy-hole subbands. The diamagnetic shift of this transition was quite small and was about the same as that of the ground state of the heavy-hole or the light-

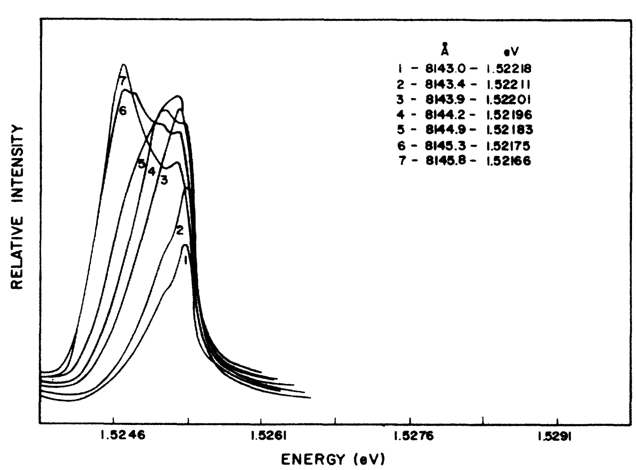

FIG. 1. Light-hole free-exciton spectra produced from PLE of the various heavy-hole free exciton components listed in the inset.

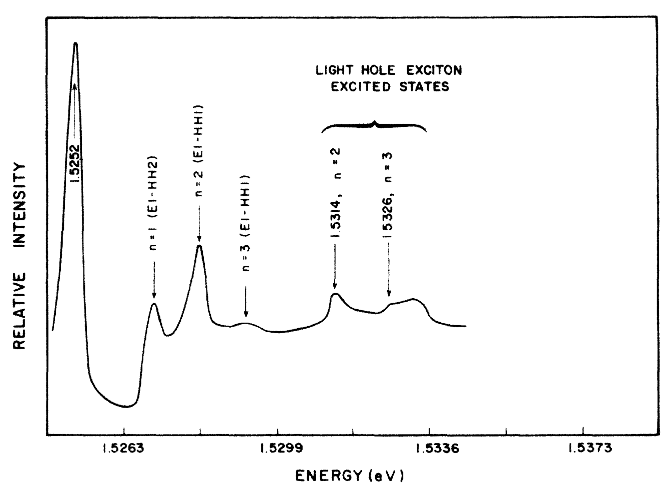

FIG. 3. The excited states of the light-hole free exciton ($E1$-LH1). This PLE spectrum is produced by measuring the relative excitation of the highest energy component of the heavy-hole free exciton.

hole exciton. This excludes the possibility that this transition is an excited state of an exciton. The diamagnetic shift of the $n=2$ state of the heavy-hole exciton ($E1$-HH1) was considerably larger than that of the $E1$-HH2 exciton and the diamagnetic shift of the $n=3$ state of the $E1$-HH1 executed was even larger than that of the $n=2$ state. The behavior of these transitions in the presence of a magnetic field is consistent with the assignments given in Fig. 2. The details of this work along with an appropriate analysis will be published elsewhere.

The transitions associated with the excited states of the light-hole free exciton are displayed in Fig. 3. The calculated values of the binding energies of the ground state ($1s$) and of the excited state ($2s$) for this well size are 7.1 and 1.35 meV, respectively.[10] This gives a value of 5.75 meV for the difference in energy between the two states which compares quite favorably with the measured value of 6.2 meV. The next higher excited-state peak observed for the free light-hole exciton is believed to contain contributions from more than one state. If one considers the onset of the peak as being the $3s$ level, its binding energy can be calculated using the same procedure described for the $3s$ state of the heavy-hole exciton. This gives a value of 0.6 meV for the binding energy of the $3s$ level. The difference between the energies of $1s$ and $3s$ states is, therefore, 6.5 meV, which compares favorably with the measured value of 7.3 meV.

In conclusion, we have observed the $2s$ and $3s$ states of both the heavy-hole exciton and the light-hole exciton in a 225-Å $Al_{0.35}Ga_{0.65}As$-GaAs MQW structure using photoluminescence excitation spectroscopy. The experimentally measured values of the differences in energies between the ground state and the excited states of both excitons agree quite well with the available calculated values as well as with estimates based on a hydrogen atom analog. The assignment of these states has been further verified from magnetic field measurements.

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