Fine-structure features due to wave-function localization in coupled GaAs-$Al_xGa_{1-x}As$ quantum wells

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(Received 21 November 1991; revised manuscript received 10 April 1992)

Photoluminescence (PL) measurements performed on nominally symmetric, coupled, double GaAs-$Al_{0.3}Ga_{0.7}As$ quantum wells grown by molecular-beam epitaxy (MBE) show fine-structure features within the observed excitonic transitions. This fine structure is interpreted in terms of wave-function-localization effects arising from deviations of the double wells from being perfectly symmetric. If the coupled-well structure is slightly asymmetric, with one well slightly larger than the other, the symmetric wave function is strongly localized in the wider well, while the antisymmetric wave function is strongly localized in the narrower well. Theory predicts that the energy spread covered by the fine-structure features resulting from the collapse of excitons associated with the symmetric combination of isolated electron and heavy-hole wave functions will be greater than that for the collapse of excitons associated with the antisymmetric combination of isolated electron and heavy-hole wave functions, in agreement with the experimental results reported here. Additionally, parity-forbidden as well as parity-allowed transitions are observed. The breakdown of the parity selection rule is believed to result from symmetry reduction arising from the presence of layer imperfections such as the clearly observed thickness variations. In some cases extrinsic transitions are also observed; magnetic-field-dependent PL measurements show these to be free-to-bound transitions (free hole to bound electron). Fine structure is also observed in the extrinsic transitions and is similar in nature to that observed for the intrinsic transitions. The asymmetry of the coupled wells is ascribed to surface kinetic phenomena occurring during MBE growth.

INTRODUCTION

It has been shown that both monolayer[1-9] and submonolayer[10,11] well-size fluctuations can be reflected in the photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy of isolated GaAs-$Al_xGa_{1-x}As$ quantum-well (QW) structures. Such fluctuations can give rise to fine-structure features within the heavy-hole free-exciton (HHFE) and light-hole free-exciton transitions, with the energy spacing between the fine-structure features corresponding to well-width fluctuations of an integral number of monolayers,[1-9] or, in some cases, corresponding to effective well-width fluctuations of less than a single monolayer.[10,11] These fine-structure features have been explained in terms of inhomogeneities within or between isolated QW's in single- or multiple-quantum-well systems. More recently, similar fine-structure features have been observed in coupled double-QW and superlattice systems.[12-14] In coupled double QW's, coupling causes the degenerate, isolated well states to mix and split into discrete states. The coupled QW states are comprised of symmetric and antisymmetric linear combinations of the envelope functions of the isolated well states. The lowest-energy excitonic transition is due to recombination of electrons and heavy holes associated with the symmetric combination of isolated well eigenfunctions. A slightly higher-energy transition is due to recombination of electrons and heavy holes associated with the antisymmetric combination of isolated well eigenfunctions.

Considering a model of two rectangular, coupled QW's of nominally equal width, Lang and Nishi[15] showed that, by introducing a slight asymmetry in well size, the ground-state wave function becomes highly localized in the wider well. Littleton and Camley[16] extended the investigation to larger numbers of coupled wells and confirmed the results of Lang and Nishi[15] for two coupled QW's, showing that the symmetric wave function is strongly localized in the wider well. They also showed that the antisymmetric wave function is strongly localized in the narrower well. They further showed that, for a given well-width difference, the energy spread of the ex-

citonic transition associated with the electron and heavy-hole symmetric states is greater than that for the transitions associated with the electron and heavy-hole antisymmetric states. In this work, PL and PLE measurements are made on nominally symmetric, coupled, double GaAs-$Al_{0.3}Ga_{0.7}As$ quantum wells grown by molecular-beam epitaxy (MBE). Fine-structure features are found within the low-lying excitonic transitions observed in PL and higher-lying excitonic transitions observed in PLE. This fine structure is interpreted in terms of wave-function-localization effects arising from deviations of the double wells from perfect symmetry. Extrinsic, free-to-bound transitions, when observed, are examined with regard to their detailed line shape and compared with those obtained for the excitonic transitions. The results are shown to be consistent with a model of inequivalent coupled wells; i.e., each double-well structure is slightly asymmetric. Specific MBE surface kinetic processes are identified that might explain such a deviation from perfect symmetry.

To aid in assigning PL and PLE transitions, transition energies are predicted using a four-band $\mathbf{k}\cdot\mathbf{p}$ model[17] that has proven accurate for both strained and unstrained systems. This model uses measured effective masses of the conduction, heavy-hole, light-hole, and splitoff bands, as well as measured energy gaps, to deduce Luttinger parameters and Kane matrix elements for each host material. The boundary conditions between adjacent layers are rigorously implemented. Both parity-allowed and parity-forbidden transition energies are calculated in order to observe selection rules operating in the PL and PLE spectra. Calculations based on a theory incorporating valence-subband mixing effects predict zero oscillator strength for parity-forbidden transitions.[18] However, nominally parity-forbidden transitions in related structures have been observed.[14,19–23]

EXPERIMENTAL ARRANGEMENT

A single sample was studied that contained 30 cycles of three different sized, symmetric, coupled GaAs-$Al_{0.3}Ga_{0.7}As$, double QW's, with nominal well widths (L_z) of 325, 200, and 100 Å. The coupled QW's were separated by 20-Å $Al_{0.3}Ga_{0.7}As$ barriers, while each pair of coupled QW's was isolated from adjacent pairs by 100-Å $Al_{0.3}Ga_{0.7}As$ barriers. The structure was grown by conventional solid-source MBE on a 2-in.-diameter, n^+GaAs substrate using a Varian Gen II MBE machine. The substrate was nominally misoriented 6° from (100) towards (111)Ga. A Perkin-Elmer cracker cell was used to produce dimeric arsenic as the arsenic growth species. The GaAs and $Al_xGa_{1-x}As$ growth rates were 0.7 and 1.0 monolayers/sec (one monolayer equals 2.8275 Å), respectively. The As_2 to Ga (uncorrected) beam equivalent pressure ratio was 14 and gave rise to an As-stabilized (2×4) surface reconstruction during GaAs growth at 580±10 °C. The buffer-layer sequence consisted of 1000-Å GaAs, followed by a ten-cycle (30-Å $Al_xGa_{1-x}As$–30-Å GaAs) superlattice, followed by 5000-Å GaAs. Grown on top of this buffer was 30 cycles of coupled 325-Å-wide wells with 20-Å $Al_{0.3}Ga_{0.7}As$ coupling barriers, followed by 30 cycles of coupled 200-Å wells, followed by 30 cycles of coupled 100-Å wells. The center of each 325-, 200-, and 100-Å well was spike doped with approximately 10^9 cm^{-2} Be acceptors as well. The acceptors were deposited during a 2-sec growth interrupt, which may have resulted in some surface smoothing. The substrate temperature during growth was 570±10 °C. This combination of growth temperature, growth rate, and substrate orientation has previously been shown to result in GaAs-$Al_xGa_{1-x}As$ heterostructures of very high optical quality.[24]

The PL and PLE were excited with an Ar^+ ion-laser-pumped tunable dye laser using Styryl 9 dye. The pump power used was approximately 1.25 W/cm^2. The measurements were made at 2 K with the sample immersed in liquid He. A magnetic field oriented perpendicular to the growth direction was used to observe diamagnetic shifts of PL transitions. The spectra were analyzed with a high-resolution 4-m spectrometer equipped with an RCA C31034A photomultiplier tube for detection.

RESULTS AND DISCUSSION

The results observed were similar for each L_z value examined and, therefore, we focus mostly on the results obtained for the 325-Å coupled wells. The PL spectra for the 325-Å coupled double QW's are shown in Fig. 1. Excitonic transitions associated with electron and heavy-hole symmetric (antisymmetric) states are labeled *S*-HHFE (*A*-HHFE). Fine-structure features with energy separations corresponding to approximately a ±3-monolayer variation in well size are observed in both the *A*-HHFE and *S*-HHFE transitions. These well-size fluctuations might be due to nearly perfectly symmectric double QW's, with each pair of wells varying slightly in width from that of the next pair, since 30 pairs of a given nominal size were grown. However, it is more likely that the double QW's are somewhat asymmetric, with one

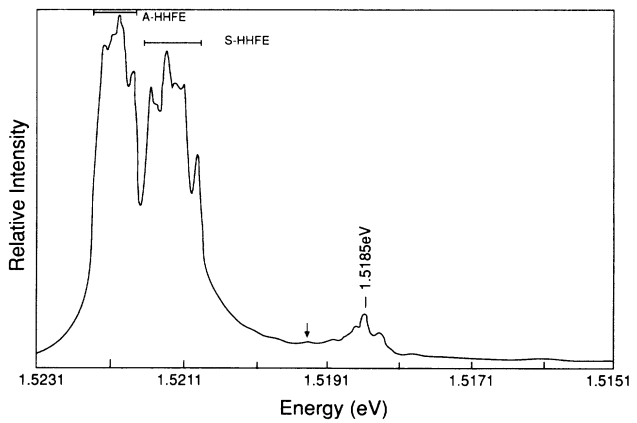

FIG. 1. PL due to *S*-HHFE and *A*-HHFE collapse for the 325-Å coupled double-QW structure with 20-Å coupling barriers. Extrinsic transitions are shown in the 1.5185-eV energy region. Fine-structure features corresponding to well-width fluctuations are seen for both the intrinsic and extrinsic transitions. The arrow marks weak extrinsic features associated with *A*-HHFE (see text).

well being slightly larger than the other.

The latter possibility would result in wave-function localization with the electron and heavy-hole symmetric wave functions localized in the wider well and having the lowest transition energy.[15,16] The electron and heavy-hole antisymmetric wave functions would be localized in the narrower well giving the next higher-energy transition. The measured energy separation between the *S*-HHFE transition and the *A*-HHFE transition is 0.9 meV, while the calculated energy separation between these two transitions is 0.5 meV. The measured energy spread of the *S*-HHFE transition is 0.67 meV, while the calculated value, assuming a ±3-monolayer fluctuation in well width, is 0.33 meV. The measured energy spread for the *A*-HHFE transition is 0.39 meV, while the calculated value is 0.24 meV. The measured and calculated energy spread of the *S*-HHFE transition is greater than that of the *A*-HHFE transition, in agreement with the predictions of Ref. 12, thereby leading additional credence to the interpretation in terms of wave-function localization. If the well-size variations occurred from pair to pair of QW's, with each pair being symmetric, the energy spread of the *S*-HHFE and the *A*-HHFE would be expected to be more nearly the same.

In the same coupled double-QW structure, extrinsic transitions are observed in the 1.5185-eV energy region. Fine-structure features are also observed in the extrinsic transitions, reflecting the well-size fluctuations observed in the intrinsic transitions. The observed transition energy suggests a free-to-bound transition (free hole to bound electron), as has been observed in isolated wells of similar size.[25] The magnetic-field dependence of both the *S*-HHFE and this extrinsic transition are shown in Fig. 2. The solid and dashed curves correspond to applied magnetic-field strengths of zero and 27 kG, respectively, with the magnetic field applied normal to the growth direction. The observed diamagnetic shift of the *S*-HHFE transition is 0.58 meV, while that of the extrinsic transition is 0.82 meV. The increased diamagnetic shift is expected for a free-to-bound transition.[25] It is likely that the highest-energy fine-structure feature on the extrinsic transition (arrow) is associated with the *A*-HHFE intrinsic transition because of the increased fine-structure spacing compared to the other fine-structure features. The reduced intensity of the extrinsic features associated with the *A*-HHFE transition suggests that the donor thermalizes rapidly compared to the time scale for emission. This is somewhat expected since the thermalization time scale is expected to be of the order of picoseconds, while the emission time scale is expected to be of the order of 1 nsec.

PLE for the 325-Å coupled double-QW structure is shown in Fig. 3. The detector hold position is 1.5213 eV, which is near the center of the *S*-HHFE transition. The calculated energies of parity-allowed and parity-forbidden transitions are designated by vertical bars at the base of the figure, with asterisks denoting parity-allowed transitions. The calculated *S*-HHFE energy was 2.0 meV lower in energy than the measured *S*-HHFE energy. To adjust for this difference all of the calculated energies are shifted in Fig. 3 to 2.0 meV higher in energy.

Fine-structure features associated with energy variations due to approximately ±1-monolayer fluctuations in well size were observed for the 200-Å coupled QW's. Four fine-structure components were observed within the *S*-HHFE transition, which covered an energy range of 0.75 meV, while the calculated energy spread for this transition is 0.67 meV. The *A*-HHFE had a measured energy spread of 0.55 meV; the calculated energy spread is 0.53 meV. Thus, this well size shows the same trend as the 325-Å coupled wells, with the *A*-HHFE transition having a smaller energy spread than the *S*-HHFE transition. This is the expected result if the fine-structure features result from wave-function localization. The observation of nominally parity-forbidden transitions is thought to be the direct result of symmetry reduction as well.

The PLE spectra (not shown) for the 200-Å coupled

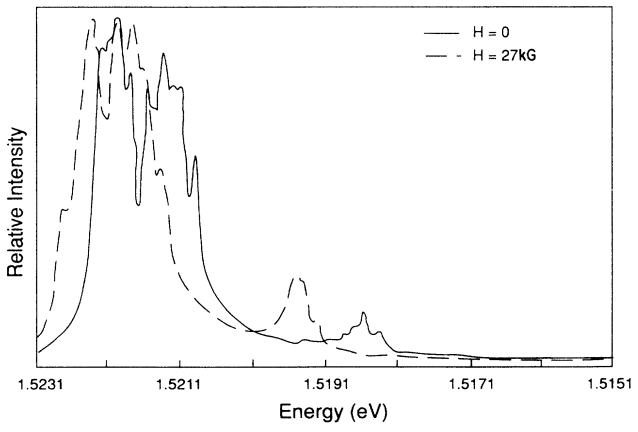

FIG. 2. PL spectra from Fig. 1 shown in zero magnetic field (solid curve) and in an applied field of 27 kG (dashed curve) applied normal to the growth axis.

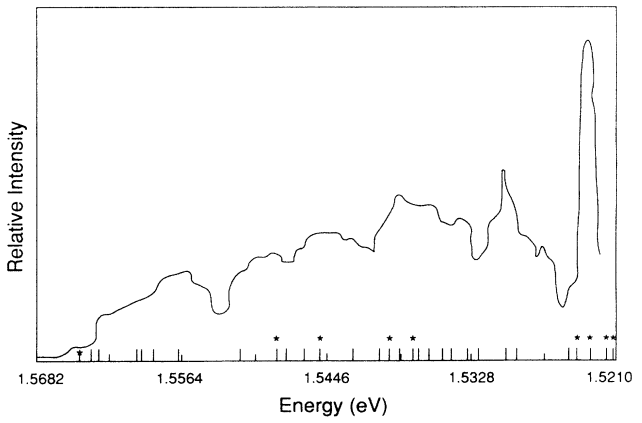

FIG. 3. PLE spectra for the 325-Å coupled double-QW structure with the detector holding at the center of the *S*-HHFE transition. Calculated energies of the transitions are shown as the vertical bars at the base of the figure. The allowed transitions are labeled with an asterisk.

QW's were found to be similar to those found for the 325-Å coupled wells (Fig. 3). Thus, a similar breakdown of parity selection rules was observed and is due to the same effects as discussed above for the 325-Å QW's. In the case of the 100-Å coupled QW's, the energy separation between the S-HHFE and A-HHFE is predicted to be about 8.0 meV, which is considerably greater than for the wider wells. Because of this large energy separation, only the S-HHFE states are populated at low temperature and thus are observable in PL. Two fine-structure features were observed in the PL spectra, corresponding to a ± 1 monolayer variation in well width. The antisymmetric states were observed in PLE but, because of increased linewidths relative to those obtained for the wider wells, these features were poorly resolved. Thus, for this size well, a comparison between the energy spread covered by the different excitonic transitions was not made. We believe, however, that the well-size fluctuations are again due to wave-function localization.

The asymmetry of the coupled-well structures is very likely due to the previously predicted[26] tendency for $Al_xGa_{1-x}As$ surfaces to increase monotonically in roughness with increasing $Al_xGa_{1-x}As$ layer thickness. Since the isolating barriers were all 100 Å in thickness while the coupling barriers were 20 or 30 Å wide, the first well grown in a double-well structure would have a rougher "inverted interface" than that of the second well.

SUMMARY

Fine-structure features were observed in the excitonic transitions in coupled double-GaAs-$Al_{0.3}Ga_{0.7}As$ QW's. These features were interpreted in terms of wave-function localization resulting from slight asymmetry in the coupled wells. The symmetric electron and heavy-hole wave funtions are strongly localized in the wider well, while the antisymmetric electron and heavy-hole wave functions are strongly localized in the narrower well. Both parity-allowed and parity-forbidden transitions were observed in these coupled QW systems. The breakdown in parity selection rules is believed due to structural symmetry reduction because of layer imperfections in the form of differing inverted interface roughness of coupled wells. Extrinsic transitions showing fine-structure features resulting from well-size fluctuations were also observed that reflect the fine-structure features of the intrinsic transitions.

ACKNOWLEDGMENTS

The authors thank J. E. Ehret and E. N. Taylor, for MBE support, and R. W. Walline and G. L. McCoy, for technical support. This work was partially supported by AFOSR. D.C.R, B.J., and P.W.Y. were supported by the USAF under Contract No. F33615-86-C-1062.

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