Low-temperature photocurrent studies of electron-state coupling in asymmetric coupled quantum wells

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We report the results of low-temperature photocurrent studies of an asymmetric coupled-quantum-well system in which the degree of asymmetry is relatively large. A straightforward interpretation of the results in terms of an avoided level crossing of electron states is obtained because the electron levels can be delocalized by application of an electric field, whereas the hole levels remain localized. The optical properties of this system are particularly sensitive to the conduction-band well depth and can therefore be used to determine band offsets.

There has been a great deal of interest recently in the optical properties of semiconductor quantum-well structures because of both the potential for practical applications and the wealth of new physical phenomena that have been discovered in these structures. Of central importance is the quantum-confined Stark effect discovered by Miller *et al.*,[1,2] in which applied electric fields on the order of 10^5 V/cm produce large red shifts in the energies of the excitonic absorptions in rectangular quantum wells.

Because of the flexibility in their designs, coupled-quantum-well systems have generated significant interest both theoretically[3,4] and experimentally,[5–9] although most of the work performed to date has been concerned with symmetric or slightly asymmetric coupled quantum wells. Low-temperature, high-resolution spectroscopic studies of symmetric[7] and slightly asymmetric[8] systems demonstrated an abundance of spectral features resulting from the breakdown of selection rules appropriate for isolated quantum wells. This spectral richness has tended to hinder the unambiguous interpretation of the observed phenomena and has therefore prevented detailed comparisons of theory with experiment in these systems.

In this paper, we report the results of low-temperature photocurrent measurements[10,11] on a GaAs/$Al_xGa_{1-x}As$ asymmetric coupled-quantum-well system in which the degree of asymmetry is relatively large. This system was examined previously[4,9] because of its unusual nonlinear optical properties. The band diagram for the system, consisting of two quantum wells of significantly different widths separated by a thin barrier, is shown in Fig. 1.

In Fig. 1(a) the magnitude of the applied bias is zero. Electron and hole wave functions are predominantly localized in either well, and the optical properties are similar to those of a pair of uncoupled quantum wells. In Fig. 1(b) the system is biased to bring the two electron levels into resonance, where strong coupling between the levels results in delocalization of both electron states, with the wave functions having substantial amplitudes in each of the wells. This coupling results in an avoided level crossing and an associated minimum energy splitting between the states that depends on the width and the height of the barrier between the wells.[12] Figure 1(c) shows the system biased beyond the point where the electron levels couple. The wave functions are again localized in separate wells, and the optical transitions regain the characteristics of isolated wells. As we show here, the field dependence of optical transitions in this system unambiguously displays the effects of the coupling between electron levels without the simultaneous hole coupling that occurs in nearly symmetric coupled-well systems.[5–8] As a result, a detailed comparison of calculations with experiment is possible, enabling, e.g., a determination of the band offsets in the GaAs/$Al_xGa_{1-x}As$ system.

The sample used in this study was grown by molecular beam epitaxy at a substrate temperature of 640° C. The quantum-well region of the sample consists of seven periods of the asymmetric coupled-quantum-well structure (9.5-nm GaAs well, 2.4-nm $Al_xGa_{1-x}As$ barrier, 4.5-nm GaAs well, and a 9.8-nm $Al_xGa_{1-x}As$ spacer layer). This undoped structure is imbedded in a *p-i-n* diode for reverse biasing. We determined the thicknesses and compositions of the well and barrier layers by measuring the energies of the $Al_xGa_{1-x}As$ absorption edge (to determine the Al mole fraction) and the lowest-energy excitonic transition in the wide well (to determine its width) using photocurrent spectroscopy under a slight forward bias (i.e., nearly flat band). These two parameters gave the absolute growth rates for GaAs and $Al_xGa_{1-x}As$, which we used along with programmed shutter times to determine all other layer thicknesses. These thicknesses and compositions are consistent with growth rates and Al mole fractions determined from calibration samples and with transmission electron microscopy measurements (of layer thicknesses) of the sample.

The sample was processed into 250-μm mesa diodes, with contacts on the top and bottom faces, and mounted in a continuous-flow, liquid-helium cold-finger cryostat. Chopped light from a tungsten-filament lamp was dispersed by a 0.75-m monochromator and focused onto one of the diodes through a window in the cryostat. A

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current-sensitive preamplifier and a lock-in amplifier were used to detect the photocurrent generated in the diode as a function of the photon energy and the bias on the sample.

Figure 2 shows the 8-K photocurrent spectra in the 1.52–1.57-eV region for three applied biases corresponding (approximately) to the conditions shown in Fig. 1. (The curves have been offset vertically for clarity.) All of the features observed in this spectral region arise from transitions involving hole states localized in the wide well. The features above 1.6 eV (not shown here) include narrow-well and excited-state wide-well transitions and show the effects of coupling between both electron and (excited-state) hole levels. Analysis of the higher-energy spectra ($\hbar\omega \geq 1.6$ eV) will be reported at a later date. Optical nonlinearities, reported previously[9] in samples of this type, were not observed in the present sample for the light intensities used in these experiments.

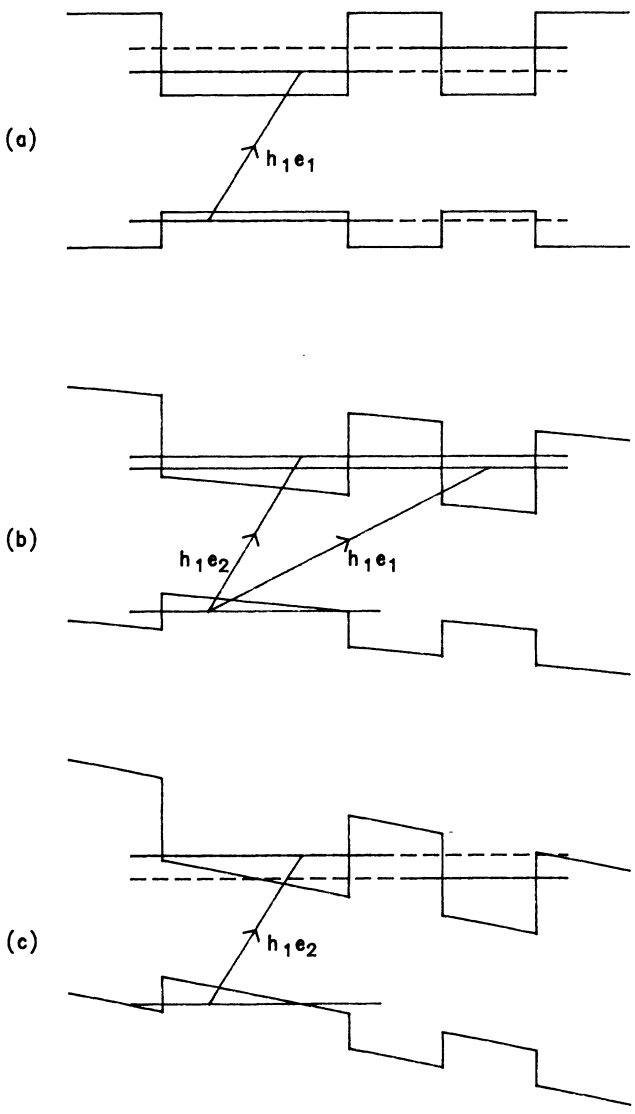

FIG. 1. Energy-band diagram for the asymmetric coupled-quantum-well system for three values of the applied bias: (a) zero bias, (b) biased to resonance, and (c) biased beyond resonance. The lowest-energy pair of electron levels (e_1,e_2) and the lowest-energy heavy-hole level (h_1) are shown; solid lines indicate regions where the wave functions are large and dashed lines where the wave functions are small. Arrows show observed electron-hole transitions. The h_1 state remains localized in the wide well for all values of reverse bias (as do the first excited heavy-hole level h_2 and the lowest-energy light-hole level l_1, not shown).

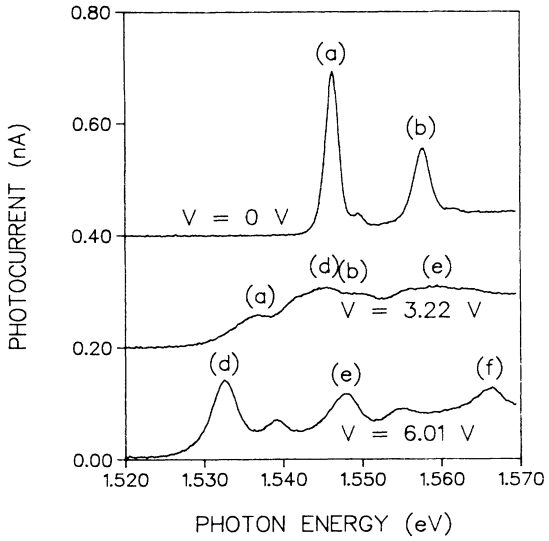

FIG. 2. 8-K photocurrent spectra of the asymmetric coupled-quantum-well sample at three values of the applied reverse bias: 0 V, 3.22 V, and 6.01 V. Transitions of interest are labeled as follows: (a) h_1e_1; (b) l_1e_1; (c) h_2e_1 (not shown); (d) h_1e_2; (e) l_1e_2; (f) h_2e_2.

At zero bias, the transitions labeled h_1e_1 and l_1e_1 are quite sharp, with a full width at half maximum of ~ 2.1 meV. At a reverse bias of 3.22 V, the coupling of the electron levels in the two wells results in significant oscillator strength in all transitions between the localized hole states (h_1, l_1, and h_2) and the pair of delocalized electron states. The transitions h_2e_1 and h_2e_2 correspond to transitions that are forbidden by symmetry for zero field in isolated wells, but are allowed for nonzero fields.[10] The dominant features in the spectrum for the 6.01-V reverse bias are the h_1e_2, l_1e_2, and h_2e_2 transitions. The small peaks on the high-energy side of each of the main features arise from the last wide well of the seven coupled-well pairs, which was ~ 0.5 nm narrower than the others due to an error in shutter timing during growth.

Figure 3 shows the peak energies of the transitions of interest as a function of applied reverse bias. The avoided level crossing associated with the interaction of electron states is seen in the h_1 and l_1 transitions for reverse biases between 2 and 4 V. Also shown in Fig. 3 are the calculated peak energies determined by a single-band envelope-function approximation, with the assumption that 70% of the GaAs/$Al_xGa_{1-x}As$ energy-gap discontinuity occurs in the conduction band. Field-independent

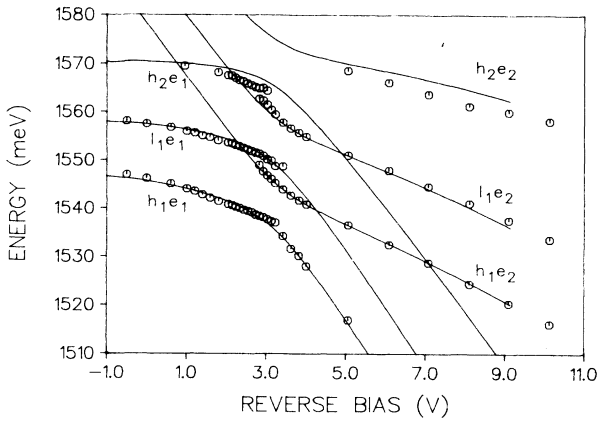

FIG. 3. Observed (⊙ symbols) and calculated (solid lines) peak transition energies as functions of applied reverse bias. In the calculations, the applied bias was determined from the electric field by using the expression $V=-1.9+0.092E$, where V is in units of V and E is in units of kV/cm (see text).

exciton binding energies of 8 and 12 meV were used, respectively, for heavy- and light-hole transitions. A linear relationship was assumed between internal electric field (the theoretical parameter) and applied bias (the experimental parameter). We determined a built-in diode potential of 1.9 V from the 77-K current-voltage characteristics of the diodes, and an effective intrinsic-region thickness of 0.92 μm by matching the calculated and measured quantum Stark shifts of the "allowed" heavy-hole transition in the high-field region, well away from resonance. This latter value is consistent with results of room-temperature capacitance-voltage measurements performed on the sample.

Figure 3 shows clearly that the observed phenomena are associated with the avoided level crossing of the electron states. The experimental values of the minimum energy splitting of electron levels and the bias voltage at which the minimum occurs are 9.4 ± 0.3 meV and 3.24 ± 0.10 V, respectively. (These were obtained by fitting the average of the splittings obtained from the "allowed" light- and heavy-hole transitions in the vicinity of the crossing, as a function of bias voltage, to a parabolic form.) The calculated splitting, 9.7 meV, and crossing bias, 3.25 V, are in excellent agreement with the experimental results. Generally, for the forbidden transitions ($h_2e_{1,2}$), the difference between calculated and observed transitions is on the order of 2–3 meV. The agreement is much better than this for the "allowed" transitions, even in the resonance region.

The calculated values for the minimum electron energy splitting and the crossing bias are strong functions of the height of the conduction-band barrier. By repeating the energy-level calculations for a number of different values of the barrier height we were able to deduce that a conduction-band offset of 70% of the energy-gap discontinuity was consistent with the experimental data. (For comparison, the calculated minimum splittings and crossing voltages are 12.4 meV and 2.13 V, respectively, for a 50% offset and 8.6 meV and 3.98 V, respectively, for an 80% offset.) Previous results for the band offset vary from 85% (Ref. 13) to 55%,[14] with the results reported most recently in the 60–65 % range.[15,16]

The highly asymmetric system described here has many advantages over symmetric (or slightly asymmetric) systems for studying the details of coupling between quantum wells. As shown in Fig. 1, it is possible to electrically tune the system completely through the resonance, which is not possible with nearly symmetric systems. The simultaneous coupling of hole states and electron states in nearly symmetric systems greatly complicates the interpretation of the absorption spectra.[7,8] As seen in Fig. 3, the use of the asymmetric well system enables the observed electronic transitions to be interpreted unambiguously as a function of applied bias. The coupling between the hole levels can be studied in a similar way by growing a structure in which the ordering of the wells is reversed relative to the direction of the field. The flexibility of design allows the crossing bias and the degree of coupling to be chosen arbitrarily, which is of potential importance in the design of optical devices using the effects of coupling and in the investigation of new phenomena unique to coupled quantum-well systems.

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