PHYSICAL REVIEW B

## Transitions to confined states of the split-off band in GaAs-(Al,Ga)As multiple-quantum-well heterostructures

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By studying the low-temperature photoluminescence excitation spectra from a number of GaAs-(Al,Ga)As multiple-quantum-well heterostructures with various well widths and Al fractions we provide the most conclusive evidence yet published for the observation of optical absorption from a confined hole state in the split-off band of GaAs to the lowest confined electron sub-band.

The optical spectra of GaAs-(Al,Ga)As quantum wells have been extensively studied, with the transitions observed being interpreted in terms of the excitonic transitions from the  $\Gamma_8$  valence band to the conduction band. In a recent publication<sup>1</sup> it was suggested that a weak feature seen in the photocurrent spectra of multiple-quantum-well samples was associated with an allowed transition from the lowest confined state in the  $\Gamma_7$  spin-orbit split-off band of the GaAs well to the lowest electron subband extremum (denoted SO1-E1). The features were observed at 55 K in two samples with well widths of 75 and 110 Å and a barrier material with an aluminum fraction close to 0.35. Spectra showing these features are given in Fig. 1. Strong allowed transitions appear as minima due to absorption in the quantum wells which reduces the photoresponse in the GaAs layers below the quantum wells.<sup>2</sup> The features could not be explained by any combination of the electrons and either light or heavy holes, and were consistent with the known spin-orbit splitting of GaAs (341 meV) (Ref. 2) and the confinement energy of the SO1 subband as estimated from the measured effectivemass values.<sup>3</sup>

In this paper we present further evidence for the existence of the SO1-E1 transition, deduced from the lowtemperature (about 8 K) photoluminescence excitation (PLE) spectra from a number of GaAs-(Al,Ga)As multiple-quantum-well (MQW) samples. In all cases a sharp peak is seen at high energy (>1.9 eV) and the strength of this feature and the variation of its energy as a function of well width and aluminum fraction provide good evidence that the peak is associated with the SO1-E1 transition. This feature, seen in all samples, cannot be identified with an energy consistent with any combination of electron and either heavy- or light-hole transitions.

The samples used in this study were grown by molecular beam epitaxy. All the layers were deposited on [001]oriented semi-insulating GaAs substrates held at a nominal substrate temperature of 650 °C. The growth sequence was (a) a 1- $\mu$ m GaAs buffer, (b) 0.13  $\mu$ m of (Al,Ga)As, (c) 60 periods of GaAs wells and (Al,Ga)As barriers, and (d) 0.13  $\mu$ m of (Al,Ga)As. None of the layers were intentionally doped. Well thicknesses and AlGaAs compositions are summarized in Table I.

Low intensity PLE spectra at about 8 K were recorded using an argon-ion pumped tunable dye laser or over an extended range using a lamp and monochromator combination. The 8-K PLE spectra of three of the samples are shown in Figs. 2(a)-2(c).

We have calculated the expected energies of the exciton transitions to help with the identification of peaks in the spectra. The calculations used an implementation of



FIG. 1. Photocurrent spectra at 55 K from two GaAs-(Al,Ga)As multiple-quantum-well samples with (a) 75-Å GaAs wells (no. 1) and (b) 110-Å GaAs wells (no. 2). In both cases the (Al,Ga)As barriers had an Al mole fraction of 0.35. The position of the SO1-E1 transition is indicated on each plot. (A full discussion of all the other spectral features is given in Ref. 1.)

7785

| Sample no.        | $L_z$ (Å) | Al fraction | Temperature<br>(K) | SO1- $E1$ (eV) |       |
|-------------------|-----------|-------------|--------------------|----------------|-------|
|                   |           |             |                    | Expt.          | Calc. |
| 1 <sup>i</sup>    | 75        | 0.35        | ~55                | 1.924          | 1.914 |
| 2 <sup>i</sup>    | 110       | 0.35        | ~55                | 1.889          | 1.883 |
| 3 <sup>ii</sup>   | 90        | 0.36        | 8                  | 1.904          | 1.905 |
| · 4 <sup>ii</sup> | 76        | 0.41        | 8                  | 1.926          | 1.923 |
| 5 <sup>ii</sup>   | 80        | 1.0         | 8                  | 1.934          | 1.932 |

TABLE I. Sample details, observed transition energies in (i) photoconductivity and (ii) photoluminescence excitation measurements and calculated transition energies, including an exciton binding energy taken to be 11 meV.

Bastard's envelope function model<sup>4</sup> for the confined electron states and an effective-mass calculation which ensured that the envelope function F and the product  $(1/m^*)dF/dz$  were continuous at the quantum-well boundaries for the split-off states.

The effective masses for electrons in GaAs and AlAs



FIG. 2. Low-temperature photoluminescence excitation spectra from multiple-quantum-well samples with (a) 90-Å GaAs wells and Al<sub>0.36</sub>Ga<sub>0.64</sub>As barriers (no. 3), (b) 76-Å GaAs wells and Al<sub>0.41</sub>Ga<sub>0.59</sub>As barriers (no. 4), and (c) 80-Å GaAs wells and AlAs barriers (no. 5) showing the positions of the SO1-*E* 1 excitonic absorption. Spectra shown over the extended range were recorded with a "lamp and monochromator" while those up to  $\sim 1.75$  eV used a tunable dye laser as excitation source. [Note: The dip in the spectrum close to 1.975 eV in (c) is due to a top cladding region of (Al,Ga)As.]

were taken as  $0.0665m_0$  and  $0.15m_0$ , respectively, and for the split-off hole bands values of  $0.14m_0$  for GaAs and  $0.27m_0$  for AlAs were chosen. For intermediate alloy compositions a linear interpolation between the two end members was made. The values of all the hole masses in GaAs and particularly in AlAs are somewhat uncertain. Experimental determinations of the split-off mass in GaAs range from  $0.133m_0$  to  $0.17m_0$  (Ref. 3) while theoretical and experimental values span the range  $0.24m_0-0.38m_0$ for AlAs.<sup>3,5</sup> The values used in the calculations are those derived by Lawaetz<sup>6</sup> from a five-band  $\mathbf{k} \cdot \mathbf{p}$  calculation of the band structures of the binary compounds.

The value used for the conduction band offset fraction  $Q_e$  was 0.67, which was determined from the energy of type-II emission from GaAs-AlAs multiple-quantum-well samples with unusually narrow GaAs layers.<sup>7</sup> Also, this value is within the range we determined earlier by fitting calculations to PLE spectra of GaAs-(Al,Ga)As multiple-quantum-well samples made by adjusting the values of  $Q_e$  and the GaAs well width  $L_z$ .<sup>8,9</sup> The potential barrier in the split-off valence band was calculated from  $Q_e$  and the known  $\Gamma$  point gaps of GaAs and (Al,Ga)As.<sup>10</sup> It was assumed that the position of the split-off band edge below the top of the valence band was given by 0.341-0.05x eV, where x is the Al fraction.

The n = 1 exciton binding energies were determined to within about 1.5 meV from a direct measurement of the 1s-2s splitting seen in the low-temperature photoluminescence of all these samples.<sup>11</sup> Slightly smaller values (1-2 meV less) were used for the n > 1 excitons in an attempt to reflect the effect of the reduced confinement.

It is common to assume that the principal peaks in the spectra are due to excitons belonging to interband transitions, where the confinement quantum numbers n of the electron and hole subbands are the same. Indeed, Brum and Bastard<sup>12</sup> have calculated a very small matrix element for the HH3-E1 transition in an envelope function approximation. This result depends on the precise relative values of the masses and well depths for the electron and hole; however, if the product  $mV_0$ , where  $V_0$  is the well depth, is the same for electrons and holes the electron and hole envelope functions for different n are mutually orthogonal. Altering the values of these parameters in this calculation enhances the result considerably.<sup>13</sup> We assumed, therefore, that some of the peaks are due to  $\Delta n = 2$ excitons. The  $\Delta n = 1$  transitions are forbidden by symmetry for zero momentum in the plane, under the assumption that the terms in the band structure linear in k are negligi7786

ble. This is no longer the case for nonzero momentum in the plane, <sup>14</sup> which implies that excitons seen in absorption must be p states or states with higher angular momentum which have a much smaller binding energy. Because these transitions are seen only when they are mixed with close by, allowed transitions, however, the exciton binding energy of the forbidden transitions and the adjacent allowed ones are expected to be quite perturbed by the interaction that causes the mixing.

In all the samples, clear peaks in the excitation spectrum on the high-energy side of the heavy-hole n=1 to electron n=1 (HH1-E1) and light-hole n=1 to electron n=1 (LH1-E1) excitons could be seen which have been identified as the 2s states of these n=1 excitons.<sup>15</sup> The observation of these peaks in the excitation spectra provides confirmation of the exciton binding energies determined from the luminescence.<sup>11</sup> In accord with the recent observations by Miller *et al.*<sup>16</sup> and by Masselink *et al.*<sup>17</sup> we see two clear peaks in the vicinity of the HH2-E2 exciton. These are associated with excitonic transitions between HH2-E2 and LH1-E2, the second of these transitions becoming allowed because of the extensive mixing of the light- and heavy-hole subbands by the electron-hole interaction. There may also be a contribution in this region from the LH3-E1 transition but the double peak feature is also seen in samples where the n = 3 light hole is not confined. The allowed transition LH2-E2 is clearly observed in all samples with an additional feature at about 1.784 eV in sample no. 3 being most probably some combination of transitions due to the parity allowed HH1-E3and HH4-E2 transitions. The same combination of allowed transitions is probably responsible for the peak at 1.865 eV in sample no. 4 but the calculations predict that the E3 and HH4 states are only just confined so it is not certain that both these transitions are present in this sample. The shoulder on the high-energy side of the LH2-E2transition in sample no. 4 is the parity forbidden HH3-E2 transition. Again it is most likely that this transition is present due to the proximity of the LH2 and HH3 confined states with the sharing of oscillator strength between the allowed and "forbidden" transitions resulting in the observation of the latter.

The energies of the features in the PLE and the photocurrent measurements which we associate with the SO1-E1 excitonic transition are shown in Table I. Also included are the theoretical values for these transitions.

The exciton binding energy for the SO1-E1 transition was estimated to be 11 meV using the calculation of Ref. 15 and assuming that the in-plane mass of the split-off hole is the same as that of the split-off hole in bulk GaAs. Comparison between the measured excitonic peaks for the SO1-E1 transition and calculated values are shown in Table I. This agreement (to within a few meV) is very satisfactory in view of the uncertainties in the effective masses, the exciton binding energies, and the exact value of the potential.

Consideration of the strength, width, and systematic dependence of this new transition on well width and aluminum fraction allows us to eliminate transitions associated with any of the light- and heavy-hole states. The strength of the transition demonstrates that we are seeing an allowed  $\Delta n = 0$  transition. The only allowed transition in this region of the spectrum of sample no. 4 would be a HH3-E3 transition. In sample no. 5, where the aluminum fraction is 1.0 the HH3-E3 transition is calculated to be shifted over 100 meV higher in energy and there is no other strong transition predicted in this region. In addition, the observation that the SO1-E1 transition is as narrow or narrower than the HH2-E2 and LH2-E2 transitions also supports our assignment since allowed  $\Delta n = 0$  transitions in quantum wells show the characteristic behavior that the excitonic linewidth increases with subband index.

In summary, the SO1-E1 transition has been observed in several multiple-quantum-well samples with differing well widths and barrier compositions making our identification unambiguous. Such a transition would be expected to make a strong contribution to the interband absorption due to its allowed nature. The transition is found to be sharp despite the fact that the confined state lies over 300 meV below the  $\Gamma_8$  valence-band edge.

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