Optical study of the electronic states of $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ quantum wells in high electric fields

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Absorption and electroabsorption spectra of (In,Ga)As/(In,Al)As quantum wells and superlattices, grown lattice matched to InP substrates, have been studied and compared with model calculations. Heavy-hole excitons dominate the spectra of uncoupled wells. They respond very sensitively to electric fields, mainly by losing oscillator strength to forbidden transitions. The ground state shows the pronounced red shift predicted from the quantum-confined Stark effect. A substantial increase of the electron mass with energy is observed. This nonparabolicity of the conduction band can be approximated by a linearly increasing mass, i.e., $m_e/m_0=0.041+0.045E/(1\text{ eV})$. Superlattices with thin barriers develop minibands whose spectra are very different from those of uncoupled wells. Features arising from M_0 and M_1 singularities of the joint density of states allow direct measurement of the width of the minibands and yield a conduction-band discontinuity of 505 ± 5 meV. With increasing electric fields, the miniband structure is altered gradually. The oscillator strength of interband transitions is moved to the center of the band at transition energies corresponding to heavy- and light-hole excitons in uncoupled wells.

I. INTRODUCTION

In quantum-well heterostructures of III-V compounds the confinement of free-carrier motion to the layer plane leads to quantized states, whose energy levels depend on the well width and on the barrier material. The resulting alteration of the electronic density of states, leading to sets of two-dimensional electron bands and enhancement of the exciton binding energy and oscillator strength, is observed in absorption and luminescence investigations. The energy states depend sensitively on electric fields, particularly strongly the ground state, which shifts rapidly to lower energy. This shift, the so-called quantum-confined Stark effect[1] has been observed in absorption experiments[2,3] and in photocurrent spectra[4] of samples containing 20–50 periods of well and barriers. Much less information exists on the behavior of higher confined states as well as of superlattice structures.

In this work we applied the electroabsorption technique to (In,Ga)As/(In,Al)As quantum wells to study the changes of the optical properties induced by an electric field with high sensitivity. This method, electroabsorption or electroreflectance, has successfully been used to unravel many details of the band structure of bulk semiconductors.[5,6] A related method, photoreflectance, which alters the field in the sample by screening with photoexcited carriers, has also been applied to quantum-well structures.[7] The system (In,Ga)As/(In,Al)As has been selected because its large barrier height of 0.5 eV for electrons[8] allows accommodation of several confined electron states. Since transitions between confined states occur below the absorption edge of the InP substrate the field-induced changes of the density of states can be measured directly in transmission. This avoids complications arising from contributions of both the imaginary and real part of the dielectric constant to the experimental spectra which are always present if working in reflectance. Line-shape analysis of electroabsorption spectra therefore is much simpler.

This paper is organized as follows. Experimental details are described in Sec. II. In Sec. III uncoupled quantum-well structures are investigated to study the impact of electric fields on the quantum-confined states. If the barriers width is small interwell coupling leads to dispersion of the electronic states in growth direction and to the formation of minibands.[9,10] The corresponding change of the two-dimensional density of states back to a three-dimensional band structure will be presented in Sec. IV. It is predicted that large electric fields will localize these band states again by shifting the energy levels of neighboring wells with respect to each other (Stark localization).[11]

II. EXPERIMENTAL DETAILS

All samples were grown by molecular beam epitaxy on *n*-type doped InP substrates at a temperature of 570 °C–580 °C. Details of growth conditions and x-ray analysis of the samples are given in Ref. 12. Periodic sequences of $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ double layers were deposited directly onto the substrate without buffer layers. A 6-nm-thick semitransparent Pt film on part of the sample served as contact to apply an external voltage with the n-type doped substrate being grounded. For the study of confined states in decoupled wells samples of large well width, 13.8 and 10.3 nm, have been chosen which accommodate several confined states. They were separated by thick barriers, 11.5 and 10.3 nm, respectively. Model calculations indicate that interwell coupling is negligible. Both samples containing 50 double layers were sufficiently thick to measure both absorption and electroabsorption spectra. For study of minibands samples with 3.5-nm-wide (In,Ga)As wells have been selected which were separated by thin barriers, 3.5 and 1.8 nm thick. Both samples contained 100 double layers.

Absorption spectra were derived from transmission measurements, accounting for multiple reflection on top and bottom faces but neglecting the small reflectance on interfaces. The light source was a tungsten-iodine lamp filtered by a 1-m grating monochromator with a bandpass of less than 1 meV. The transmitted intensity I and its change ΔI, induced by square voltage of about 1 kHz frequency, were measured with a nitrogen-cooled Ge detector followed by a lock-in amplifier to analyze the modulated part. The samples were immersed in superfluid He for rapid dissipation of Ohmic heat at large fields.

The field-induced change $\Delta\alpha$ of the absorption constant is easily obtained from the ratio of the relative change of the transmitted intensity and the thickness d of the epilayers:

$$d\,\Delta\alpha = -\Delta I / I \ . \tag{1}$$

Field-induced changes of the reflectivity are negligible because of the small reflection on internal interfaces whereas in regions of high reflectance, the metal contact and the doped substrate, the field is very small and not modulated.

The width of the minibands was calculated by a modified Kronig-Penney model.[13] For calculation of the energy states in decoupled quantum wells we used an eight-band envelope function[14] with nonparabolic electron mass and the boundary conditions of Bastard.[15] Tunneling resonance calculations were used to evaluate the shift of the confined states and their broadening due to the finite lifetime in an electric field.

III. RESULTS AND DISCUSSION: NONCOUPLING QUANTUM WELLS

A. Model calculation of the quantum-confined Stark effect

An electric field causes an energy shift of the transitions between quantum-confined states. A large red shift of the lowest confined state, exceeding by far the Stark shift in atoms or molecules, has been verified in several experiments.[2–4] A model calculation of this effect is helpful in understanding the experimental spectra. Figures 1 and 2 show the result of tunneling resonance calculations for 13.8-nm-wide wells, using the parameters in Table I and following the procedure by Klipstein *et al.*[16]

In an electric field electrons will tunnel through the barrier and the confined states degenerate to resonances which are obtained as maxima of the ratio S of amplitudes of the wave function inside and outside the well, normalized to the tunneling current. Figure 1 shows such resonances for heavy holes. The ground state moves rapidly with increasing field to lower energy but higher confined states behave differently. In low fields they all move to higher energy but, at sufficiently high field, they return back to lower energies again. Higher confined levels need larger field strength to enforce a red shift. Because of their large mass, tunneling of heavy holes is slow and the resonances remain very sharp even at fairly large fields. Some broadening is noticeable for the higher

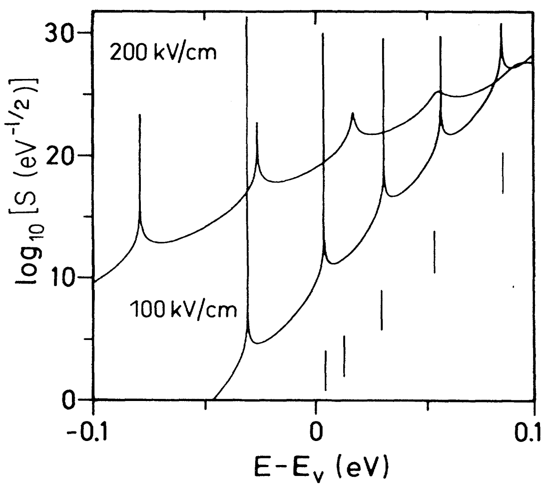

FIG. 1. Result of tunneling resonance calculations, showing the shift of the heavy-hole states in 13.8-nm-wide wells. The bars indicate the energy levels for the field-free case.

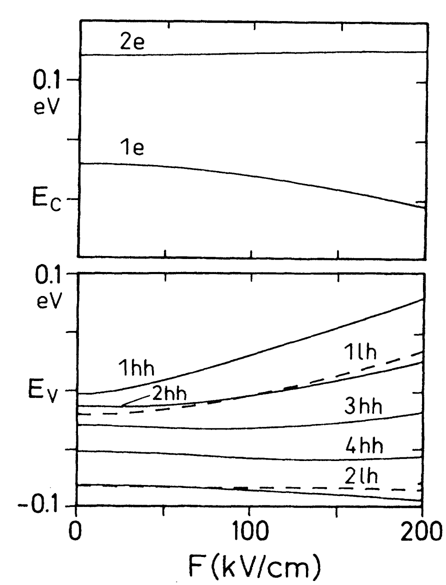

FIG. 2. Field-induced shift of some confined states of electrons and holes in 13.8-nm-wide wells.

TABLE I. Parameters used in modeling the energy states in (In,Ga)As/(In,Al)As quantum wells.

	E_g (eV)	m_e/m_0	m_{lh}/m_0	m_{hh}/m_0	$\Delta E_c/\Delta E_v$
(In,Ga)As	0.805	0.041	0.052	0.50	70/30
(In,Al)As	1.509	0.075	0.095	0.57	

confined states. Similar results are obtained for the electrons. Although, owing to their smaller mass, they tunnel faster, their resonances too remain well defined except for high-lying states close to the top of the well. Linewidth broadening due to tunneling seems negligible for the range of fields reached in our experiments.

A summary of the shift of the energy levels is given in Fig. 2. The ground states of electrons and holes shift rapidly to lower energy. The shift of the other energy levels is smaller and to opposite direction for fields less than 50 kV/cm. Complicated behavior is expected for light-hole states which cross, in certain field ranges, heavy-hole states.

B. Comparison of absorption and electroabsorption spectra

The absorption spectrum of 50 double layers of 13.8-nm (In,Ga)As and 11.5-nm (In-Al-As) at 2 K is shown in Fig. 3. Because of high doping (Burstein-Moss shift) the InP substrate was transparent up to the gap of the barrier material at 1.51 eV. The evaluation of the absorption constant from transmission is based on the thickness of the absorbing well material only, assuming negligible extension of wave functions into the barrier material. The spectrum reveals five steps, accentuated by excitons, which are related to five confined states of the electrons. Heavy-hole excitons dominate, whereas light-hole excitons give much smaller contributions. The linewidth of

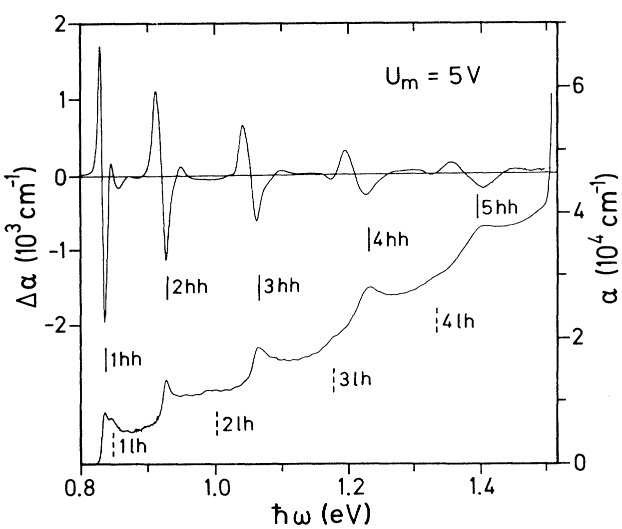

FIG. 3. Absorption and electroabsorption spectra (large modulation) of 13.8-nm-wide wells with 11.3-nm-broad barriers. $T=2$ K, modulating voltage $U=5$ V.

the exciton peaks is much larger than indicated by tunnel resonance calculations and arises from alloy fluctuations of the ternary (In,Ga)As well material[17,18] and from one monolayer well width fluctuations which affect higher-energy levels more.

Solid and dashed bars indicate allowed transitions for heavy- and light-hole excitons, respectively. The electron mass in Table I allows only for four confined states and leads to larger transition energies. Nonparabolicity of the electron mass calculated by a Kane model, involving the valence bands and the lowest conduction band, moved the energy levels slightly down but insufficient to agree with experimental data. Extrapolation of the energy dependence reported from cyclotron resonance measurements,[19] on the other hand, gave too low transition energies and predicted even a sixth confined electron state. Good agreement, within a few meV, to measured transition energies of all heavy-hole excitons in all samples was obtained for a linearly increasing electron mass:

$$m_e/m_0=0.041+0.045E/(1\text{ eV}) . \quad (2)$$

From Shubnikov–de Haas oscillations in 8.2- and 3.5-nm-thick quantum wells[20] electron masses of $0.049m_0$ and $0.053m_0$, respectively, have been deduced which, within the quoted accuracy of these measurements by $\pm 0.005m_0$, agree with the values derived from Eq. (2).

The transition energies of light holes point also to a substantial increase of their mass at higher energies. For the small mass in Table I only three confined states are predicted and the third light-hole exciton would be 40 meV higher in energy. Such a shift corresponds to an increase of the light-hole mass to $0.08m_0$. An even larger mass ($0.14m_0$) is required to adjust the fourth light-hole exciton energy.

Application of a voltage of 5 V in reverse direction leads to quite large changes with $\Delta\alpha$ up to 2000 cm^{-1} corresponding to transmission changes of more than 10%. The electroabsorption spectrum too is dominated by heavy-hole excitons which all show signals of similar line shape and also of similar strength, if the increasing width at higher energy is considered. The negative peak of $\Delta\alpha$ coincides with the corresponding peak of the absorption spectrum for all heavy-hole excitons, providing an accurate determination of the transition energies. This is particularly interesting in the case of single well structures where absorption is too weak to be directly measured, whereas the change of transmission $\Delta T/T$ is still well above the noise level ($\approx 10^{-6}$).

The leading positive peak of all features of the $\Delta\alpha$ spectra points to a red shift of all heavy-hole (hh) excitons by the electric field as expected for the 1hh exciton but not for the higher exciton states. For an estimated field of 40 kV/cm all higher confined states either move very little, resulting in a small signal, or shift to higher energy which would lead to an opposite sign of $\Delta\alpha$. Obviously the energy shift predicted from resonant tunneling calculations cannot explain the line shape of the electroabsorption spectrum involving higher confined states.

Light-hole (lh) excitons respond weakly to the electric field with line shapes quite different from the response of heavy-hole excitons. The first light-hole exciton, 1lh→1e, responds to an electric field with a small positive peak at the energy of its absorption peak, which indicates that oscillator strength is gained and dominates the predicted red shift. The unexpected behavior arises probably from mixing of the light-hole and the closely neighbored second heavy-hole sublevel.[21,22] Excitation from this state to the electron ground state, 2hh→1e, becomes allowed in an electric field and this increase of oscillator strength apparently determines the shape of the spectrum in this range. No response is resolved from the second light-hole exciton whereas the third and fourth show small negative peaks at the energy of corresponding features in absorption.

C. Electroabsorption spectra with small modulation voltage

More details of electroabsorption spectra are resolved in measurements with small modulation (±0.5 V), superimposed by dc bias to alter the field in the sample. Corresponding spectra of a sample with 50 double layers (In,Ga)As and (In,Al)As, each layer 10.3 nm thick, are shown in Fig. 4. When a small forward bias (+1.5 V) reduces the built-in field, two signals related to the first and second heavy-hole excitons are observed pointing again to a red shift with increasing field. Because of residual strain of about 2×10^{-3} in the (In,Ga)As layer of this particular sample, light- and heavy-hole exciton are almost degenerate as strain-induced splitting of the valence band compensates the different hole confinement energies. Both signals decrease with further increasing forward bias and disappear at about +3 V. They reappear at even higher forward bias in the spectrum, however with the reverse sign, indicating that the flat band position has been passed.

If by reverse bias the internal field is increased the signal becomes larger and the 1hh exciton shifts to lower transition energy as expected. Additional features develop, most clearly seen for the highest reverse bias, −9.5 V. The line shape of the signal from the 1hh exciton is altered as its negative peak partially overlaps with the light-hole exciton 1lh. The degeneracy of 1hh and 1lh observed in this strained sample at low field is lifted because the light-hole state shifts at a slower rate (see Fig. 2). Even more changes are observed in the region of the 2hh exciton. A peak appears at 0.92 eV where model calculations indicate a transition from the 3hh state to the electron ground state, 3hh→1e. An even stronger peak grows at 0.97 eV, shifting to lower energy with increasing field. Transition energy and shift support its assignment to a transition from the heavy-hole ground state to the second electron level (1hh→2e). These transitions, forbidden for the field-free case, gain strength when electric field breaks the selection rules. The negative peak at 1.0 eV, which corresponds to the second heavy-hole exciton, has shifted slightly to higher energy as expected from calculation. The different line shape of its response in Fig. 3 is obviously related to the large modulating voltage which obscures the gradual changes of the electronic states.

The dependence of electroabsorption spectra on bias voltage shows umambiguously that electric fields cause considerable increase of the strength of forbidden transitions at the expense of the allowed ones. This effect is clearly seen in model calculations of the transition probabilities (Fig. 5). We assume for simplicity infinite barrier height and neglect Coulomb interaction, following the paper of Matsura and Kamizato.[23] Without the electric field the steplike spectrum of a two-dimensional density of states is obtained where the subband spacing is largely due to the larger confinement energy of electrons. The shape of this spectrum is considerably altered by a field of 50 kV/cm (dashed line). As expected the absorption

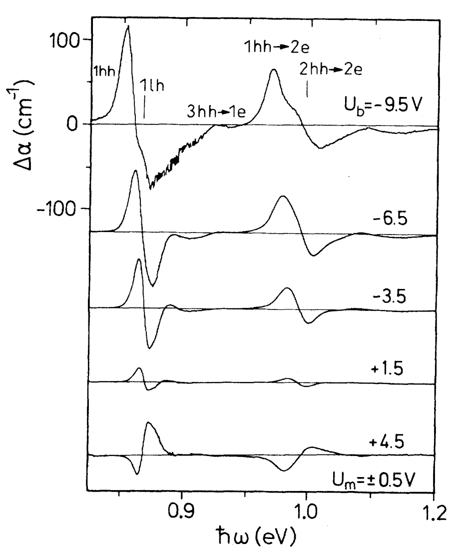

FIG. 4. Variation with dc bias of the electroabsorption spectrum measured with small modulation. Well and barriers 10.3 nm, T=2 K.

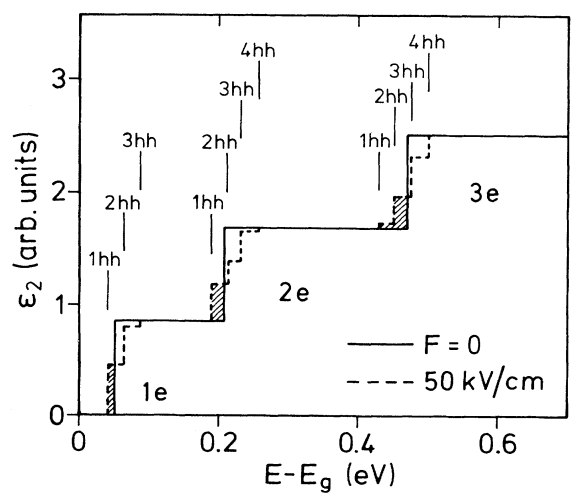

FIG. 5. Model calculations of the absorption spectrum of 13.8-nm-wide wells related to transitions from heavy-hole to electron sublevels. Excitonic effects are neglected. The solid line refers to the field-free case, the dashed line shows the absorption in a field of 50 kV/cm.

edge, related to the transition $1h \rightarrow 1e$, has moved to lower energy and its strength has decreased due to the smaller spatial overlap of electron and hole envelope functions with applied electric field. Most of the oscillator strength is acquired by the forbidden transition $2h \rightarrow 1e$ and the rest is shifted to $3hh \rightarrow 1e$. Similar changes occur for transitions to the second and third electron sublevel. The red shift of these edges, however, has a different origin. The second threshold is related to the forbidden transition $1h \rightarrow 2e$ which gained much of the strength of the allowed transition $2h \rightarrow 2e$ which has slightly shifted to higher energy and lost most of its oscillator strength. With contributions from a further "forbidden" transition, $3h \rightarrow 2e$, the absorption again reaches full strength. Such redistribution of oscillator strength is observed for all sublevels and is responsible for the similar line shape of all features of the electroabsorption spectra: a negative peak at the energy of allowed transitions, led by a positive peak at the low-energy side. For the first sublevel the positive peak indeed corresponds to a red shift of the energy states, whereas in all other cases the line shape reflects the transfer of oscillator strength from allowed to forbidden transitions. At very high field the two-dimensional density of states disappears and the spectrum will correspond to that of a three-dimensional solid in a high field showing the familiar Franz-Keldysh effect of a parabolic band edge. This relationship of quantum-confined Stark effect and Franz-Keldysh effect has been treated by Schmitt-Rink, Chemla, and Miller.[24]

The field-induced red shift of the heavy-hole exciton measured by stepping the field with dc bias agrees well with the prediction from the quantum-confined Stark effect. A comparison of experimental data, the shift of the heavy-hole exciton in an electric field, and of tunnel resonance calculations is given in Fig. 6. Quantitative agreement is obtained when we assume that 50% of the external bias voltage drops off over the quantum wells while the rest is lost in serial resistance. Large serial resistance is also indicated by the observation that 3-V forward bias is needed to compensate the internal field. The internal potential should not be larger than 1.5 V, corresponding to the gap of the barrier material.

IV. RESULTS AND DISCUSSION: COUPLED QUANTUM WELLS

A. Strong coupling

As the barrier width decreases interwell coupling overcomes the confinement of carriers to single layers and leads to evolution of minibands. Spectra of two samples have been studied, both containing 100 wells of 12 monolayers (In,Ga)As (3.5 nm), but of different barrier width, 1.8 and 3.5 nm, respectively (Fig. 7). The narrow wells accommodate only one confined state for light holes and electrons, a second confined electron state may exist just at the edge of the continuum, the conduction band of the barrier material (In,Al)As. In coupled wells of such thin barriers this electron state should merge with the continuum states.

Kronig-Penney calculations with the parameters from Table I indicate even for the sample with thin barriers, six monolayers only, that heavy holes are still fairly confined. Their bandwidth is less than 4 meV whereas both light holes and electrons develop about 125-meV-wide bands. The absorption spectrum of this sample is very different from that of decoupled wells shown in Fig. 3 or in Ref. 12. Absorption increases smoothly above 0.93 eV without the sharp edge, characteristic for the two-dimensional density of states of decoupled wells. Pronounced excitonic features are also absent but a kink at 0.97 eV indicates some fine structure. The smooth increase of absorption ends in a small peak at 1.07 eV which is followed by a plateau, typical for a two-dimensional density of states. A second absorption edge

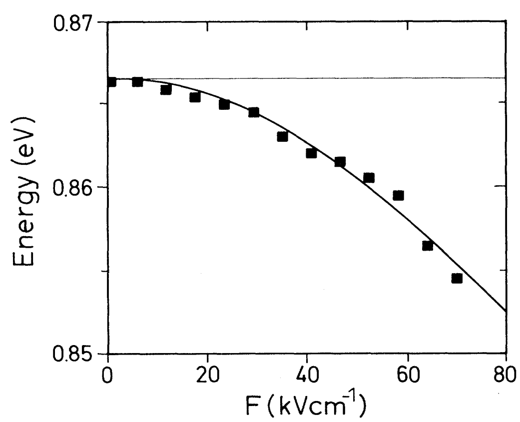

FIG. 6. Red shift of the heavy-hole exciton in 10.3-nm-wide wells. The curve, calculated by the resonant tunneling method, agrees well with data points from Fig. 4 under the assumption that 50% of the externally applied voltage change the field in the sample.

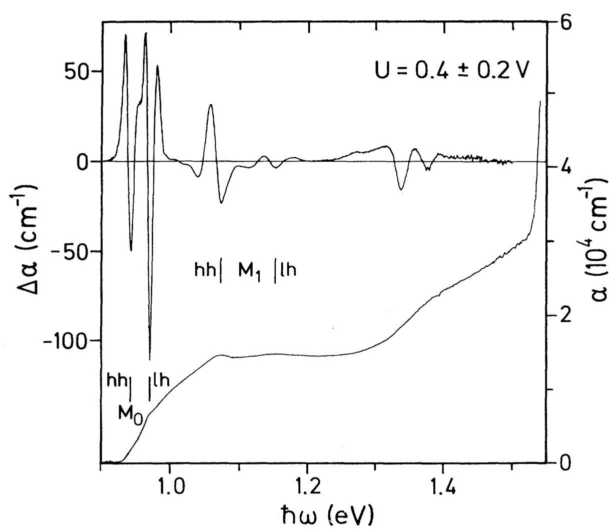

FIG. 7. Absorption and low-field electroabsorption spectrum of a superlattice with 3.5-nm-wide wells and 1.8-nm-thick barriers. $T=2$ K.

begins at 1.3 eV and slightly above 1.5 eV the absorption of the barrier material sets in.

The absorption edge resembles a three-dimensional density of states extending about 140 meV beyond the gap. Assignment of its weak features seem quite arbitrary. Fortunately, all features respond sensitively to an electric field as shown by their low-field electroabsorption spectrum, measured with small modulation (±0.2 V) on top of a small forward bias (+0.2 V). At the absorption threshold and at the kink halfway up the edge two strong signals of narrow linewidth are observed. Their line shape, the response of each transition consisting of two positive and a negative peak, is distinctly different from that of a quantum-confined Stark effect, but in perfect agreement with the Franz-Keldysh effect at a three-dimensional van Hove singularity of type M_0.[25] Similar spectra have been observed in electroabsorption spectra of (InGa)(AsP) heterostructures.[26] Both spectra show Coulomb enhancement from electron-hole interaction which deepens the negative peak. The position of these negative peaks at 0.946 and 0.977 eV gives the energy gaps between the minibands of electrons and heavy and light hole, respectively. We neglect the exciton binding energy which is small because the excitons are no longer confined.

A further pronounced signal is observed above 1.05 eV which when inverted has a line shape similar to that of the first two transitions. Such inverted line shape is predicted for a M_1 singularity, a saddle point as it must occur at the upper boundary of the minibands. We assign this signal to a transition from the narrow heavy-hole band to the top of the electron band. In the frame of the Franz-Keldysh effect at saddle-point singularities, the gap lies at the high-energy side of the spectrum, close to its last negative peak but excitonic effects may modify this assignment.

There has been some controversy on the existence of saddle-point excitons.[27,28] More recent calculations,[29] using Stahl's polariton approach, support their existence for a wide range of parameters. The well-resolved peak at 1.07 eV, marking the transition from a three- to a two-dimensional density of states, points to some Coulomb enhancement. Saddle-point excitons have large binding energy,[28] comparable to that of confined excitons, which may explain why this peak is much stronger than the Coulomb enhancement of the M_0 singularities. The absorption peak at 1.07 eV coincides with the last negative peak of the electroabsorption spectrum which overlaps with oscillations related to a further transition at higher energy. At 1.152 eV we observe a hump in the absorption spectrum and a negative peak of the electroabsorption spectrum. We assign these features to the saddle-point singularity of the light-hole miniband. It should be mentioned that similar weak peaks have been observed in photocurrent spectra of GaAs/(Al,Ga)As superlattices and were assigned to saddle-point excitons.[30]

The assignment of the various features of absorption and electroabsorption spectra to singularities of the joint density of states allows the evaluation of the width of the minibands, neglecting the small width of the heavy-hole band. We presume that the first M_0 singularity at 0.946 eV is related to the heavy hole and derive 130 and 48 meV width of electron and light-hole minibands, respectively. The width of the electron band agrees well with the Kronig-Penney calculation, 124 meV for a mass of $0.041m_0$ or slightly less if the energy dependence of the electron mass is taken into account. The width of the light-hole band is smaller than anticipated which is probably related to the rapid increase of the light-hole mass at higher energy, observed already for transitions in decoupled wells.

The absorption edge above 1.3 eV is similar to the fundamental edge and shows also a kink near 1.38 eV. Electroabsorption spectra in this range reveal a doublet with negative peaks at 1.338 and 1.377 eV. Their splitting by 39 meV is similar to that of the two M_0 singularities of the minibands. We therefore assume that both transitions are related to the hole minibands and a higher electron state. The only final state of appropriate energy is the conduction band of the barrier material. From an energy of 1.338 eV of the transition from the heavy-hole state to the conduction band we derive a barrier height for electrons of 502 meV, assuming the gap of (In,Ga)As to be 0.805 eV and a confinement energy of 31 meV for the heavy hole. This leads to a valence-band discontinuity of 202 meV and a ratio $\Delta E_c/\Delta E_v = 71/29$ for the band offsets which is in good accordance with the ratio 70/30 reported from *C-V* measurements.[11] A conduction-band discontinuity of 0.44 eV, obtained from fitting three transitions at room temperature[31] is too small to be compatible with our results.

These values are based on an assignment of the first M_0 singularity to the heavy-hole state. The alternative, attribution of this singularity to the light-hole miniband, can be ruled out. Such assignment leads to miniband widths of 100 and 109 meV for electrons and light holes, respectively. The width of the electron band is compatible with results from a Kronig-Penney model but the light-hole band seems too wide. Furthermore, the heavy-hole band would be located within the light-hole band and additional effects from band mixing are expected. Additional objections arise from considerations of the second absorption edge to continuum states above 1.3 eV. The heavy-hole state would be related to the second transition at 1.377 eV, leading to barrier heights of 541 and 163 meV for electrons and holes, respectively. Such a low valence-band barrier is too small to accommodate the four light-hole states observed in 13.8-nm-wide wells.

B. Intermediate coupling

Coupling of confined states decreases rapidly as the barrier width increases. Figure 8 shows absorption and electroabsorption spectra of the superlattice with the same well width as the previous sample but twice the barrier width (3.5 nm). Kronig-Penney calculations predict 26- and 43-meV-wide bands for electrons and light holes, respectively, and less than 1 meV for the heavy hole. The absorption edge starts at 0.97 eV and rises steeply up to 1.01 eV showing again a kink near 0.98 eV. A further slow increase leads to a broad peak near 1.07 eV. The

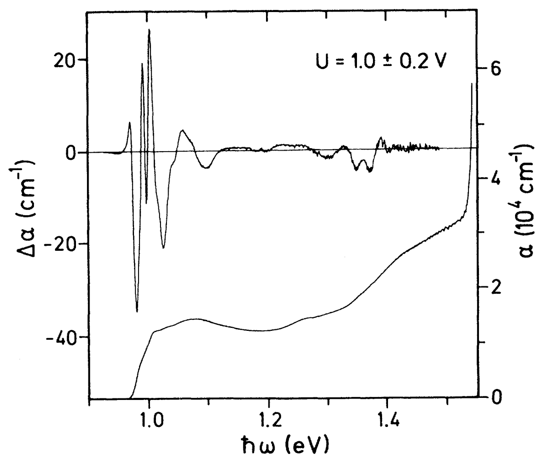

FIG. 8. Absorption and electroabsorption spectra of a superlattice with 3.5-nm-wide wells and 3.5-nm-thick barriers. $T=2$ K.

second absorption edge to continuum states begins above 1.3 eV. The absorption spectra of both superlattices are very similar, differing mainly by the width of the steep part of the edge which is reduced to 35 meV as compared to 140 meV in case of narrow barriers. A similar contraction of the bandwidth is observed in electroabsorption. The spectral shape is very complicated and depends sensitively on temperature and field strength, an indication that the response of several transitions overlap. The first negative peak at 0.974 eV is attributed to the M_0 singularity of the heavy hole and electron. By analogy to the former example we assign the absorption peak at 1.010 eV on top of the edge and its corresponding feature in the electroabsorption spectrum to the M_1 saddle point of the electron band. Both features overlap with the response of the band gap of the light hole and electron which is assigned to the second negative electroabsorption peak at 0.997 eV, slightly above the kink in the absorption edge. The light-hole saddle point M_1 is probably related to the weak signals above 1.05 eV and the broad absorption peak at 1.07 eV. The resulting bandwidth is 36 meV for the electrons in fair agreement with the calculation. The width of 37 meV for the light-hole band is only an estimate because of the uncertain determination of its upper band edge.

The splitting of heavy- and light-hole band edges by 23 meV is also observed in the doublet above 1.3 eV related to the second absorption threshold. These transitions are found at 1.347 and 1.370 eV, very close to the values observed for the other sample and confirming the assignment to the electron continuum. The much smaller bandwidth for this case of thicker barriers rules out that the first transition belongs to the light hole. We derive an electron barrier height of 509 meV, which is very close to the barrier height of 502 meV found before.

C. Stark localization

Large electric fields F shift the energy of equivalent states in neighboring wells by $W=eFD$, where D is the period of the lattice. The degeneracy of confined states is lifted and coupling should decrease. The electrons become localized to fewer wells as the field increases and finally the confined exciton states should reappear near the center of the former minibands.[11] In addition, charge-transfer (CT) excitations, interwell transitions, are expected, where electrons and holes reside in different wells. They should appear on both sides of the intrawell excitations, equally spaced by the energy W. Transitions related to this Stark ladder have been reported from luminescence[32] and electroreflectance measurements[33] GaAs/(Al,Ga)As superlattices.

The transformation of minibands to more localized states is shown in Fig. 9 for the sample with narrow wells. Bias voltage again establishes a field in the sample which is modulated by a small square voltage, ±0.2 V, to obtain the corresponding electroabsorption spectra. The spectrum on top, obtained with small reverse bias of −0.2 V, agrees with that obtained for small forward bias (Fig. 7). The field is too small to affect the minibands and their M_0 and M_1 singularities. Changes of the line shape occur for larger fields. Reverse bias of −1 V reduces the response of the first M_0 singularity by a factor of 2 and a new feature appears near its negative peak. Similar changes are observed throughout the spectrum. Strong alteration of the spectrum is seen for larger reverse bias which leads to complex spectra with many oscillations (−1.8 and −2.6 V). As these oscillations appear the signal of the first M_0 singularity (0.94 eV) becomes weaker and that related to the light hole (0.97 eV) seems to disappear. The spectrum becomes simpler again for even higher reverse voltage, −3.4 V. It is now dominated by two strong positive peaks at 1.00 and 1.045 eV pointing to new transitions growing with increasing fields. Model

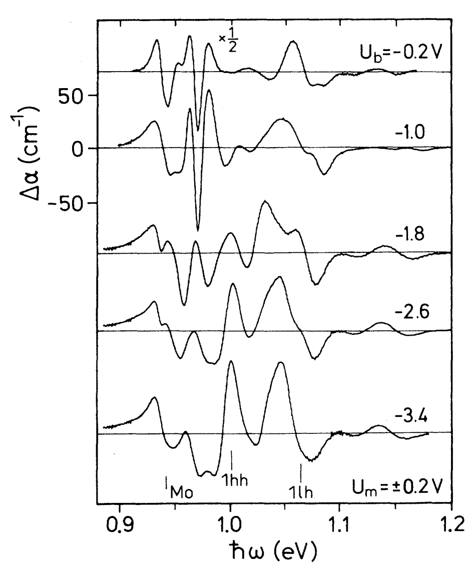

FIG. 9. Variation with dc bias of electroabsorption spectra measured with small modulation, showing the transformation of the superlattice to confined states. Same sample as in Fig. 6. $T=2$ K.

calculations for decoupled wells as well as the absorption spectrum of a sample with the same well width but wide barriers[12] indicate that these transition energies correspond to confined heavy- and light-hole excitons. The prounounced peaks have numerous sidebands which are probably related to CT excitations, the expected Stark ladder. We estimate for this spectrum a field of 50 kV/cm which shifts the energy levels in neighboring wells by 26 meV. The large number of peaks, their width, and rapidly changing line shape, however, make it difficult to follow the shift with increasing bias.

The appearance of the new strong features in the middle of the minibands proves that coupling between the wells begins to break down in high fields. This change does not occur abruptly. The spectrum still contains contributions from the M_0 singularities of the minibands. The first positive peak of the whole spectrum, related to the transition between heavy-hole and electron minibands, has not shifted with increasing field but loses strength. Electric fields thus do not reduce the width of the minibands but they reduce gradually the strength of the transitions near the absorption edge and enhance the absorption due to states in the center of the bands. The absorption edge does not shift. Similar to the case of confined exciton states the main effect of the electric field is the redistribution of oscillator strength. Although in all cases W, the field-induced splitting of levels in neighboring wells was much smaller than the width of the electron band, the onset of Stark localization is clearly seen in electroabsorption. Larger fields could not be applied because of the low resistivity of our sample. Larger fields should be possible by replacing the Pt contact by a p-type doped InP layer.

V. CONCLUSIONS

It has been shown that electroabsorption provides a simple yet very sensitive and versatile method to study quantum wells and superlattices. The spectra, related only to the imaginary part of the dielectric constant are much easier to interpret than electroreflectance spectra where contributions of field-induced changes of real and imaginary parts interfere. The method is capable of distinguishing confined and unconfined states and has sufficient sensitivity to investigate single wells. It is not limited to low temperature; spectra of similar strength and shape are observed at room temperature although thermal broadening obscures some details.

Strong derivativelike spectra are obtained from heavy-hole excitons in decoupled wells with the negative peak of $\Delta\alpha$ corresponding to the transition energy. The lowest confined states ($1\text{hh} \rightarrow 1e$) respond to the field by a large red shift which is well described by the quantum-confined Stark effect. Somewhat unexpected is the very similar line shape and strength of electroabsorption spectra involving higher confined states. Their response to an electric field is not due to their small energy shift but arises from a transfer of oscillator strength from allowed to forbidden transitions. Light-hole excitons respond weakly to an electric field with spectral line shape which depends on well width. Their response seems to involve coupling to heavy-hole states, probably by dispersion parallel to the interface.

The numerous confined states, resolved in electroabsorption spectra of wide wells, allows better modeling of their transition energies. For (In,Ga)As/(In,Al)As we found that the electron barrier is 500 meV high. The transition energies involving higher confined states indicate considerable energy dependence of the electron mass of (In,Ga)As. We find a linear increase $m_e/m_0 = 0.041 + 0.045E/(1\ \text{eV})$ from evaluation of heavy-hole excitons. The spectra point also to a rapid increase of the light-hole mass at higher energy.

Electroabsorption spectra of distinctly different line shapes are observed in samples where coupling across narrow wells leads to formation of minibands. These minibands respond by a Franz-Keldysh effect of their M_0 and M_1 singularities. The spectra allow a fairly accurate determination of the width of minibands and show how rapidly this width is shrinking if interwell coupling is reduced by thicker barriers. In the case of intermediate coupling and smaller bandwidth, contributions of several critical points of the density of states merge, giving rise to complicated spectra whose line shape alters rapidly with increasing field. At higher energy a doublet in the electroabsorption spectra reveals a second absorption threshold which corresponds to transitions from the hole minibands to the electron continuum, the conduction band of the barrier material (In,Al)As. It enables direct determination of the electron barrier height. A value of 505 ± 5 meV has been derived, which corresponds to a ratio of band discontinuities $\Delta E_c / \Delta E_v = 71/29$.

High electric fields localize the states of minibands and the quantum-confined states reappear, corresponding to intrawell transitions and accompanied by a number of other transitions which probably are related to interwell transitions. In the range of fields accessible in our study the M_0 singularity of the electron miniband has neither disappeared nor shifted in energy, in spite of these new transitions, but its absorption strength has decreased. Like in decoupled wells the dominant effect of the field is the redistribution of oscillator strength, piling up the strength of miniband transitions at the middle of the transition range.

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[1]D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. B **32**, 1043 (1985).
[2]J. S. Weiner, D. A. B. Miller, D. S. Chemla, T. C. Damen, C. A. Burrus, T. H. Wood, A. C. Gossard, and W. Wiegmann, Appl. Phys. Lett. **47**, 1148 (1985).
[3]D. A. B. Miller, J. S. Weiner, and D. S. Chemla, IEEE J. Quantum Electron. **QE-22**, 1816 (1986).
[4]K. Yamanaka, T. Fukunaga, N. Tsukada, K. L. I. Kobayashi, and M. Ishii, Appl. Phys. Lett. **48**, 840 (1986).
[5]*Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1972), Vol. 9.
[6]M. Cardona, *Modulation Spectroscopy* (Academic, New York, 1982).
[7]U. K. Reddy, G. Ji, T. Henderson, H. Morkoç, and J. M. Schulman, J. Appl. Phys. **62**, 145 (1987).
[8]R. People, K. W. Wecht, K. Alavi, and A. Y. Cho, Appl. Phys. Lett. **43**, 118 (1983).
[9]R. Tsu and L. Esaki, Appl. Phys. Lett. **22**, 562 (1973).
[10]R. Dingle, in *Festkörperprobleme*, edited by H. -J. Queisser (Vieweg-Verlag, Braunschweig, Germany), Vol. XV, p. 21.
[11]J. Bleuse, G. Bastard, and P. Voisin, Phys. Rev. Lett. **60**, 220 (1988).
[12]W. Stolz, J. C. Maan, M. Altarelli, L. Tapfer, and K. Ploog, Phys. Rev. B **36**, 4301 (1987).
[13]H. S. Cho and P. R. Prucnal, Phys. Rev. B **36**, 3237 (1987).
[14]M. F. H. Schuurmans and G. W. t' Hooft, Phys. Rev. B **31**, 8041 (1985).
[15]G. Bastard, Phys. Rev. B **24**, 5693 (1981).
[16]P. C. Klipstein, P. R. Tapster, N. Aspley, D. A. Anderson, M. S. Skolnik, T. M. Kerr, and K. Woodbridge, J. Phys. C **19**, 857 (1986).
[17]E. F. Schubert, E. O. Göbel, Y. Horikoshi, K. Ploog, and H. J. Queisser, Phys. Rev. B **30**, 813 (1984).
[18]W. Stolz, J. Wagner, and K. Ploog, J. Cryst. Growth **81**, 79 (1987).
[19]R. J. Nicholas, C. K. Sarkar, L. C. Brunel, S. Huan, J. C. Portals, M. Razhegi, J. Chevrier, J. Massies, and H. M. Cox, J. Phys. C **18**, L427 (1985).
[20]W. Stolz, J. C. Maan, M. Altarelli, L. Tapfer, and K. Ploog, Phys. Rev. B **36**, 4310 (1987).
[21]G. Bastard and J. A. Brum, IEEE J. Quantum Electron. **QE-22**, 1625 (1985).
[22]G. D. Sanders and K. K. Bajaj, Phys. Rev. B **35**, 2308 (1987).
[23]M. Matsura and T. Kamizato, Phys. Rev. B **33**, 8385 (1985).
[24]B. S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989).
[25]Y. Hamakawa, P. Handler, and F. A. Germano, Phys. Rev. **167**, 709 (1968).
[26]K. Satzke, G. Weiser, R. Höger, and W. Thulke, J. Appl. Phys. **63**, 5485 (1988).
[27]C. B. Duke and B. Segall, Phys. Rev. Lett. **17**, 19 (1966).
[28]E. O. Kane, Phys. Rev. **180**, 852 (1969).
[29]I. Balslev, Solid State Commun. **52**, 351 (1984).
[30]R. H. Yan, R. J. Simes, H. Ribot, L. A. Coldren, and A. C. Gossard, Appl. Phys. Lett. **54**, 1549 (1989).
[31]J. S. Weiner, D. S. Chemla, D. A. B. Miller, T. H. Wood, D. Sivco, and A. Y. Cho, Appl. Phys. Lett. **46**, 619 (1985).
[32]E. E. Mendez, F. Agullo-Rueda, and J. M. Hong, Phys. Rev. Lett. **60**, 2426 (1988).
[33]P. Voisin, J. Bleuse, C. Boude, S. Gaillard, C. Alibert, and A. Regrency, Phys. Rev. Lett. **61**, 1639 (1988).