Exciton mixing in the magnetophotoluminescence excitation spectra of shallow strained $\text{In}_x \text{Ga}_{1-x} \text{As/GaAs}$ quantum wells

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We report highly resolved photoluminescence excitation spectra of excitons in intrinsic pseudomorphic In, $Ga_{1-x}As/GaAs$ quantum wells (QW's). The QW parameters are chosen such that only the $n_z = 1$ electron and hole subbands are confined and the light-hole —heavy-hole exciton splitting exceeds the exciton binding energy. By using circularly polarized light the fine structure of excitonic transitions is well resolved in the spectra of quantum wells at magnetic fields $H \leq 8$ T. We observe a strong mixing of lightand heavy-hole excitons which causes optical transitions into high-angular-momentum exciton states and results in strong anticrossing effects. Two excited states of the exciton located at \approx 1 and 2.5 meV above the 1s light-hole exciton state have been observed and are related to exciton states with the carrier-wave-function delocalized into the GaAs barriers.

I. INTRODUCTION

Since the pioneering work of $\text{Dingle}, \cdot \cdot \text{excitons}$ in semiconductor quantum wells (QW's) have been subject to numerous studies motivated by the pronounced effects of quasi-two-dimensionality in the excitonic features of optical spectra. The quantities of interest are energies and oscillator strengths of the optical transitions associated with the excitation or recombination of exciton states.

Experimentally, excitons in intrinsic GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ QW's have been studied extensively by different methods including photoluminescence and photoluminescence excitation.¹⁻¹⁴ Excitonic properties in the presence of an external magnetic field are of special interest, and provide information about the excitonic binding energies and coupling of excited exciton states. 4^{-9} In particular, exciton transitions forbidden in a simple two-band model,^{4,12} and anticrossing behavior of excited exciton states, $8,9,13,14$ have been observed in magnetic field.

A large number of theoretical studies of the QW exciton in a magnetic field have been published based on a 'simple model of a two-dimensional hydrogen atom.^{2,1} Later it was shown that the complex valence-band structure and strong coupling of light and heavy holes in GaAs/Al_xGa_{1-x}As QW's results in many peculiarities in the excitonic spectrum.¹

In our paper we present photoluminescence (PL) and photoluminescence excitation (PLE) spectra of excitons in shallow strained $In_xGa_{1-x}As/GaAs$ QW's. The strain-induced splitting of the valence band in these QW's nearly coincides with the valence-band offset. The question of the energy position of the light-hole band in these QW's (above or below the GaAs valence band) is still under discussion. This peculiar position of the light-hole subband is expected to result in an additional resonance between excited excitonic states in the barrier and in the QW . 10,21 Here we will show that experimental studies of shallow QW's of high quality can reveal these effects and allow us to follow their disappearance with increasing QW depth. We have presented highly resolved photoluminescence excitation spectra of excitons in undoped shallow strained $In_xGa_{1-x}As/GaAs$ quantum wells which contain only one bound electron and hole subband state $(n_z=1)$. Due to the internal strain of the In_xGa_{1-x} As QW layer, the light-hole-heavy-hole exciton splitting in this QW exceeds the exciton binding energy. This allows us to study the coupling of the light- and heavy-hole exciton states in detail.

The paper is organized as follows. The experimental details are described in Sec. II. In Sec. III, PL and PLE spectra of excitonic states in narrow $In_xGa_{1-x}As/GaAs$ QW's in magnetic fields $H < 8$ T are described in detail. The magnetic-field behavior of exciton transitions including light and heavy holes is discussed in Secs. IV and V. In Sec. IV we analyze a strong mixing of light- and heavy-hole excitons which results in the appearance of optical transitions into high angular momentum exciton states, and in strong anticrossing effects. In Sec. V we discuss the nature of two additional excited states of the exciton located near (\approx 1 and 2.5 meV above) the 1s light-hole exciton state and relate them to states with wave functions which are strongly delocalized into the GaAs barriers (QW-confined quasi-three-dimensional excitons).

II. EXPERIMENT

The measurements were performed on undoped $\text{In}_{x}\text{Ga}_{1-x}\text{As}/\text{Ga}$ As heterostructures with a single QW or several QW's of different thickness grown by solid source molecular-beam epitaxy (MBE) on a GaAs substrate.

QW's with $x=0.033$ and 0.045 and thicknesses in a wide range $(2-10 \text{ nm})$ were investigated. The samples were immersed in pumped liquid helium in a cryostat equipped with a superconducting coil. Both PL and PLE spectra were recorded with the use of a tunable Ti-sapphire laser and a double-grating monochromator. The experiments were performed in the Faraday configuration, with σ^+ and σ^- helicities of the exciting light. The emission was detected by a cooled photomultiplier.

III. PL AND PLE SPECTRA

The conduction band of the shallow and rather narrow In_xGa_{1-x} As QW's investigated here consists of only one $n_z = 1$ subband, whereas the valence band consists of two, heavy hole (hh) and light hole (lh), $n_z = 1$ subbands. Representative PL and PLE spectra for the 5-nm QW at zero magnetic field are shown in Fig. 1. The emission spectrum shows only one strong and rather narrow line (the linewidth at half maximum is ≈ 0.5 meV) due to the recombination of the heavy-hole exciton in the ground state which we label as lshh. Here the first number indicates the main quantum number, s corresponds to the zero angular momentum of the hole envelope function, and the letters hh are used to indicate that we deal with excitons with a heavy hole.

In PLE measurements the intensity was detected at the half maximum of the low-energy side of the Ishh emission line. The PLE intensities are expected to reflect the absorption spectra. The high quality of the QW permits observation of a well-resolved narrow line 2shh at the ¹ .505 eV, which is due to the transition into the 2s state of the hh exciton. Higher-energy hh exciton transitions are not resolved. The energy gap between 1s and 2s states of the hh exciton is ≈ 6.3 meV, in qualitative agreement with calculations.²³

Figure 1 also displays strong and very narrow lines in the spectral range $1.507 - 1.511$ eV, corresponding to the energy range of the lh transitions. The line at ¹.5072 eV energy range of the lh transitions. The line at 1.5072 eV
is due to the excitation of the 1slh excitons.¹¹ Only this line is well resolved in QW's with rather large potential fluctuations. The gap between this line and two others

FIG. 1. PL and PLE spectra of a 5-nm ${\rm In}_{0.045}{\rm Ga}_{0.955}{\rm As/GaAS}$ QW at 1.8 K.

marked A and B is about $2-3$ meV. The investigation of the transition energies for QW's with different thicknesses and an In content $x=0.045$ reveals a rather weak dependence of the energy gaps between the lines labeled \vec{A} and \vec{B} and the 1slh transition. It increases by less than ¹ meV when the QW thickness changes from 3 to 7 nm. With increasing QW width the intensity of the lines ^A and B decreases, and for QW's with widths larger than ¹0 nm line A is not resolved at the high-energy side of

the 1slh line, whereas line B transforms into a rather

FIG. 2. PLE spectra of a 5-nm $In_{0.045}Ga_{0.955}As/GaAs QW in$ different magnetic fields normal to the QW plane in σ^- polarization. (a) Magnetic-field strength $H \le 1.32$ T. (b) Magneticfield strength $H > 1.5$ T.

weak hump at the edge of the band-to-band transitions, as observed earlier in Ref. 11. The energy gap between transitions 1slh and A or B is much smaller than the energy difference between the ground and excited states for a quasi-2D exciton in type-I GaAs/Al_xGa_{1-x}As QW's. Therefore the peculiar structure of the valence band in strained $\text{In}_{x} \text{Ga}_{1-x} \text{As/GaAs}$ seems to be responsible for this unusual behavior of excited states of the lh excitons.

Additional information about the excitonic states is obtained from the magnetooptical spectra. Figures 2(a) and 3(a) present the PLE spectra of a 5-nm $In_{0.045}Ga_{0.955}As/GaAs QW$ in the range of small magnetic fields $H = 0.33 - 1.32$ T recorded in the σ^- and σ^+ po-

FIG. 3. PLE spectra of a 5-nm $In_{0.045}Ga_{0.955}As/GaAs QW$ in different magnetic fields normal to the QW plane in σ^+ polarization. (a) Magnetic-field strength $H \le 1.32$ T. (b) Magneticfield strength $H > 1.5$ T.

larization, respectively. The behavior of the magneto-PLE spectra at higher fields is illustrated in Figs. 2(b) and 3(b), respectively. As shown in Figs. 2 and 3 the PLE spectra in magnetic field reveal a very rich structure. First of all, Figs. 2(a) and 3(a) show that the allowed transitions to the states nshh $(n=2-4)$ are already well resolved both in σ^+ and σ^- polarization at fields as low as 0.3 T.

With increasing magnetic field the lines due to different transitions shift to higher energy and we observe various crossings and anticrossings between different transitions. For example, the nshh emission lines cross the emission line lslh connected with the lh exciton recombination without any indication of the interaction of the nshh and 1shh states. This is well seen in Fig. 3(b) at $H = 2-2.33$ T, when the 2shh line moves through the lslh line, or in Figs. 2(a) and 3(a) at $H = 0.66-1.1$ T when there occurs the crossing of the 3shh and 1slh lines. The absence of interaction of these transitions is due to different symmetries of the nshh and islh states. Note that in contrast, we observe a strong interaction, for example, between a structure occurring at 1.5076 eV for $H = 0.88$ T, while the transition moves close to the 1slh line (1.5084) eV). Both lines exchange oscillator strengths and show a pronounced anticrossing behavior in the transition energies for a magnetic field of ≤ 1 T.

A comparison of the spectral positions of the nshh lines in σ^- and σ^+ polarizations in Figs. 2 and 3 shows that the spin splitting for these states is smaller than the line half-width. As expected for such transitions, they show rather regular fan charts for energies well below the energy of the lslh exciton state.

The fan charts of the absorption peaks obtained with σ^- and σ^+ polarization are shown in Figs. 4 and 5, respectively. In addition to the nshh lines, there are several additional lines marked $nd^{-(+)}$ that appear in spectra with increased magnetic field slightly below the islh transition. In contrast to the nshh lines, these lines display well-pronounced anticrossings with the lslh excitons. Figure 2(b) illustrates the anticrossing of the $3d^-$ and

FIG. 4. Magnetic-field dependence of the exciton states excited with σ^- polarization for a 5-nm-thick QW, and H normal to the QW plane.

FIG. 5. Magnetic-field dependence of the exciton states excited with σ^+ polarization for a 5-nm-thick QW and H normal to the QW plane.

lslh lines that is most pronounced. The anticrossing of the 1slh and $3d^+$ lines is shown in Fig. 3(a) at $H=0.55-1.1$ T. In Fig. 2(a) one can also find the anticrossing of the $4d^-$ and 1slh lines at $H \approx 1$ T, and that of the $5d^-$ and 1slh lines at ≈ 0.5 T. As can be seen from the magneto-PLE spectra in Figs. 2 and 3, after anticrossing the nd lines acquire the properties of the lslh emission, whereas the original 1slh line decreases in intensity and disappears from the spectrum when the resonance conditions are disturbed. This means that the coupling of the lslh state with excited hh states marked nd causes the appearance of otherwise forbidden transitions of hh excitonic states, i.e., states with high angular momenta. 22 A comparison of the spectra in Figs. 2 and 3, respectively, shows that the states appearing in the σ and σ^+ polarization are different from each other, and that the interaction is most pronounced for the states active in the σ^- polarization.

The behavior of lines A and B also shown in Figs. 2 and 3 differs markedly from that of the 1slh line. Line A splits into two well-separated components active in different polarizations, but none of them shows any marked interaction both with the nshh or nd lines. Line B also splits into two components active in different polarizations. Note that in contrast to the 1slh line these components show well-pronounced anticrossing with the nshh lines rather than with the nd lines. This indicates that the 1slh states and line B are of different symmetries.

IV. COUPLING OF THE 1slh EXCITON WITH EXCITED ndhh STATES OF hh EXCITONS

The theoretical consideration of the influence of valence-band mixing on excitonic states was carried out by Bauer and Ando. 22 To indicate the symmetry of an exciton (consisting of an electron and hole from the $n_z = 1$ subband), a notation of (n, m) was used. Here *n* is the number of the Landau level, and m is the angular momentum number $(m = ..., -2, -1, 0, 1, 2, ...)$ denotmomentum number $(m = \ldots, -2, -1, 0, 1, 2, \ldots)$ denoted as $m = \ldots, d^-, p^-, s, p, d, \ldots$, respectively. In magnetic field the label $3d^-$ corresponds, for example, to the transition involving the second hole Landau level and the zero electron Landau level. The label $3d^+$, on the other hand, indicates a transition involving the second electron and the zero hole Landau level. The calculation of Ando and Bauer predicts that the most pronounced interaction occurs for $3d^-$ hh and 1slh excitonic states of Γ_{6g} symme try. This coupling was recently found in magneto-optical studies of narrow GaAs/Al_xGa_{1-x}As QW's.⁸ It appears in the PLE spectra with σ^- polarization.

Figures 2(b) and 4 show that the anticrossing of $3d$ ^{-hh} and "ls"lh states in our QW occurs at magnetic fields $H = 1.7-4$ T. Indeed, it can be seen from Fig. 2(b) that at $H=1.7$ T a line corresponding to the 3d⁻hh transition appears slightly below the allowed 2shh transition. This behavior is consistent with the results of Ando and Bauer.²² The intensity of this line increases strongly when approaching the energy of the lslh exciton with increasing field. In the same field range ($H \approx 1.7$ T) the 1slh emission line at 1.509 eV begins to move strongly to higher energies and its intensity decreases monotonically. With increasing H the original $3d$ hh line reaches the spectral position of the 1slh exciton [see data for $H=5.5$ T in Fig. 2(b)] and can again be considered as ^a "ls"lh If in Fig. 2(0)] and can again be considered as a state
term with the hole angular momentum $M = \frac{1}{2}$. In partic ular, the comparison of Figs. 4 and 5 shows that at $H = 6-7$ T the energetic position of this transition in the σ^- polarization again nearly coincides with the "1s"lh term in the σ^+ polarization. In contrast to the strong interaction observed, e.g., for the $3d^-$ and 1slh transition Figs. 2(a) and 4 show no interaction between 2shh and 1slh transitions. This is due to the different symmetry of these states (*nshh*: Γ_{7g} , 1slh: Γ_{6g}), which prevents an interaction independent of the helicity of the exciting $light.²²$

In addition to the $3d$ ^{-hh} – "1s"lh state coupling, one finds in Figs. 2-S many more of smaller strength. First we note the coupling of the "1s"lh with the $3d⁺$ hh state [compare Figs. 3(a) and 5]. Different from the $3d$ ^{-hh} state interaction, this coupling appears in the σ^+ polarization.²² Because the $3d^+$ state involves the third electron Landau level and the first-hole Landau level, 22 the transition occurs at higher energies than the $3d$ ⁻ transition discussed above. Therefore we observe an anticrossing behavior of the $3d^+$ state and the "1s"lh already at small fields of the order of 1 T. The $3d⁺$ hh emission line appears in the spectrum in Fig. 3(a) slightly below 3shh at \approx 0.7 T, and reaches its maximum intensity indicating the change of its character to 1slh already at 1.5 T. As shown in Fig. 5 for higher magnetic fields, the variation of this transition versus H is quite similar to that of 1shh. The increased magnetic field results in a diamagnetic shift of this line to higher energies. The magnitude of the shift is a little smaller than that for the lshh transition. This is connected with a larger value of the in-plane mass for the lh state.

Let us now consider the coupling of the lslh magnetoexcitons with higher hh states which are clearly distinguished in Fig. 2(a) at $H<1.5$ T. These are again most pronounced in the σ^- polarization. Due to symmetry considerations, a strong coupling of the 1slh states $(M = \frac{1}{2})$ is expected only with nd ⁻hh states.²² Therefore we assign the emission line appearing at 0.77 T near the position of the 3shh state to the $4d$ ⁻hh transition. With increasing magnetic field this line grows in intensity due to its change into 1slh. But, as seen from Figs. 2 and 4, a very strong coupling of this term with the approaching $3d$ hh results in its further transformation to the $3d$ hh never being of real "1s"lh character. At last, a similar situation takes place at $H \approx 0.5$ T, when there occurs an interaction of the "1s"1h state with the approaching $5d$ hh state.

As clearly can be seen, e.g., in Figs. 2 and 4, we observe a strong dependence of the level repulsion on the difference in the value of n . The repulsion between the nd ⁻hh and "ls" ih states decreases monotonically with n, in accordance with the calculations.²² Figure 4 shows that it is \approx 3 meV for $n = 3$, 0.6 meV for $n = 4$, and only 0.2 meV for $n = 5$, and is not observed for larger *n*. In the σ^+ -polarized spectra the repulsion of the nd⁺hh and 1slh states is significantly smaller than discussed above for nd hh states. It is equal to ≈ 0.4 meV for the $n = 3$ state, and it is not resolved for states with larger n.

V. QW-CONFINED EXCITED QUASI-THREE-DIMENSIONAL (3D) EXCITONIC STATES

In addition to the hh and 1slh exciton transitions discussed in Sec. IV, Fig. ¹ shows two more pronounced lines denoted A and B . To determine the symmetry of the corresponding excitonic states, we refer to the fan charts. Being of the same symmetry as 1slh, all the higher nslh transitions have to display qualitatively the same behavior as 1slh. Figures 2 and 3 show that neither line A nor B interacts with, e.g., nd ⁻hh states. Therefore these lines can be assigned to nslh excitons. Indeed, Figs. 2 and 3 show that line A , located 1 meV above the ground state, is split in magnetic field into two (σ^+ and σ^-) components. We found no marked coupling for both of these components, either with nshh states of Γ_{7g} sym metry or with *ndhh* and mixed *ndhh*-1slh states of Γ_{6g} symmetry. The upper excited state interacts with 2shh and 3shh states both in the σ^+ and σ^- polarizations. This implies that this state is of Γ_{7g} symmetry and therefore cannot be related to the nslh states, which are of Γ_{6g} symmetry.

One more argument against connecting lines A and B with $ns(n \geq 2)$ exciton states is their relatively small diamagnetic shift $\Delta_{dia,i}$. It can be seen from Fig. 5 that for both A and B components $\Delta_{dia, A(B)} \approx \Delta_{dia, 1s}$, and simultaneously it is several times smaller than $\Delta_{dia, 2shh}$.

In principle, besides nslh states, parity allowed optical transitions (due to the lower symmetry of the QW) are possible from the even two-dimensional (2D) lh excitonic states with high angular momenta. However, they are also to be excluded from the consideration. In addition to the sma11 diamagnetic shift, mentioned above, which is not expected for such transitions, there are two more arguments that deal with states not related to 2D-ndlh transitions.²⁴ Figure 1 shows that lines A and B have a relatively high oscillator strength comparable to that of the 1slh state. Furthermore, Fig. 3 indicates that none of the A and B transitions has any additional splitting in magnetic fields intrinsic to high angular momentum states. The magnetic field splits these lines into doublets with one component active in σ^+ polarization and the other active in σ^- .

In order to explain transitions A and B , we most likely have to consider states for which the wave functions are significantly delocalized into the GaAs barriers. As mentioned above, the QW is too shallow to have any excited, confined free-electron or free heavy-hole states. However, such a confinement is possible when the Coulomb interaction is included. There are several arguments to associate states A and B with exciton states whose hole (or electron) is located mainly in the GaAs barriers. First, the half-width of lines A and B is very small. As shown in Fig. 1 the half-width of lines A and B is smaller than the half-width of the lslh transition by a factor of 2. This indicates a reduced inhuence of alloy fluctuations and surface roughness, and would be consistent with the participation of only one 2D level in the optical transition.

The second argument for an assignment of the A and B transitions is based on a comparison of the diamagnetic shift for different states in the magnetic field normal (Δ_{dial}) and parallel (Δ_{dial}) to the QW plane. Figure 6 shows the variation of the different transition energies in a magnetic field oriented parallel to the quantum-well plane. In this geometry the interaction of the transitions is strongly reduced and we observe different diamagnetic shifts of the transitions. It can be seen from Figs. 5 and 6 that for hh excitons Δ_{dial} is markedly smaller than Δ_{dial} . This effect is connected with an enhanced particle confinement in the z direction by the QW potential. Thus the ratio of Δ_{dial} to Δ_{dial} is equal to \approx 2 for the 1shh state. It increases nearly two times for the 2shh state because of the larger size of the 2shh wave function in the QW plane. In contrast, for lines A and B the ratio Δ_{dial} to $\Delta_{\text{dia} \parallel}$ is close to unity, which indicates a quasi-3D charac ter for the wave functions. Therefore we interpret lines

FIG. 6. Magnetic-field dependence of the exciton states for a 5-nm-thick QW and H parallel to the QW plane.

 A and B as due to transitions involving excitons formed by an electron (or hole) bound in the $In_xGa_{1-x}As$ QW and a hole (electron) delocalized into GaAs barriers. This interpretation is consistent with the small PLE linewidth, the strong enough PLE intensity, and the small anisotropy of the diamagnetic shift.

The determination of the symmetry of the considered excitonic states is possible from their mixing with other states. Figure 5 shows a well-pronounced anticrossing of line 8 with 2shh and 3shh transitions that indicates that it involves a state of Γ_{7g} symmetry. The lowest excitonic state with this symmetry is expected to be the state with a hole wave function having two nodes in the x direction (similar to that noted as the h_{13} state in Ref. 22). Note however, that it is hardly reasonable to classify states delocalized into the GaAs barriers as 1h or hh states. Line A does not show any significant mixing with other excitonic states. That does not allow us to say much about the structure of the corresponding exciton state.

The interpretation shown above has to answer the question of why the considered QW-confined quasi-3D exciton states with high angular momenta are located so near to the transition energy of the 1slh state. This can be explained if we take into account that the 1h subband in the strained $In_1Ga_{1-x}As/GaAs$ QW's lies slightly below the valence band in GaAs (type II). Therefore the confinement of the hole in the lslh exciton is already strongly determined by the Coulomb attraction of the electron localized in the In_xGa_{1-x} As layer. A repulsive potential created by an increased light-hole energy in the In_xGa_{1-x} As layer would result in a warping of the hole density in the z direction (normal to the QW plane) and, hence, in a decreasing gap between the nodeless ground state and the excited QW-confined states with additional nodes. Thus the excitonic states originating from high angular momentum 3D excitons oriented along the z direction are expected to have a relatively small energy separation from the lslh state, and a relatively weak diamagnetic shift for H normal to the QW plane. The states of 2p symmetry are dipole forbidden; however, those of 3d symmetry are expected to have a high enough oscillator strength to be observable in absorption.

VI. CONCLUSION

We have presented highly resolved photoluminescence excitation spectra of excitons in undoped shallow strained $In_xGa_{1-x}As/GaAs$ quantum wells which contain only one bound electron and hole subband state $(n_z = 1)$. Due to the internal strain of the $In_xGa_{1-x}As$ QW layer, the light-hole —heavy-hole exciton splitting in this QW exceeds the exciton binding energy. This allows us to study the coupling of the lh and hh exciton states in detail. Using circularly polarized light we were able to resolve the fine structure in the PLE spectrum of quantum wells with and without magnetic field. We observe a strong mixing of lslh excitons with excited states of the hh exciton with angular momentum $m = 2$ (nd^{\pm}hh excitons) which results in the appearance of optical transitions into high angular momentum states, and in strong anticrossing effects.

Two additional, excited states of the QW-confined quasi-3D excitons with unexpectedly small energy gaps between them and the 1slh state of \approx 1 and 2.5 meV were found. These energies are markedly smaller than the energy of the 2s excitons in type-I QW's. The transitions have a symmetry different from the Γ_{6g} symmetry intrinsic to nslh QW states. Their small energies are attributed to the type-II alignment of light-hole states in the strained $In_{x}Ga_{1-x}As/GaAs$ QW's, which results in a delocalization of the hole wave function into the GaAs barriers. This is consistent with the observed symmetry, the weak anisotropy of the diamagnetic shift of these lines, and their narrow PLE linewidth.

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- 24Though the optical transitions from gerade excitonic states with any angular momenta are not parity forbidden, their oscillator strength is very low, as the angular momentum has to be scattered in the photon absorption act. As a consequence such transitions are usually not observed in optical spectra. None of them is observed in our QW for the hh exciton states, until it borrows oscillator strength from the coupling with the 1slh exciton.