Dependence of the binding energy of the acceptor on its position in a GaAs/$Al_xGa_{1-x}As$ quantum well

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Transitions from the ground state, 1*s*, to the first excited state, 2*s*, of the Be acceptor confined in GaAs/$Al_xGa_{1-x}As$ quantum wells (QW's) have been observed via two independent spectroscopic techniques: (a) two-hole transition of the bound exciton (BE) observed in selective photoluminescence, and (b) resonant Raman scattering. The transition energy has been determined from such measurements for acceptors with positions varying from the center to the edge of the QW using δ-doped samples. The results obtained are found to agree well with recent theoretical calculations for acceptors close to the center but disagree when approaching the edge of the QW. The binding energy of the BE is found to decrease when the acceptor position shifts from the center, where the binding energy has its maximum value towards the minimum at the QW interface.

INTRODUCTION

Variation of the binding energy of an acceptor bound hole, confined in a quantum well (QW), with the impurity position has been calculated in several approximations. The original calculation by Bastard[1] assumed a hydrogenic impurity and infinite barrier height. More realistic calculations, where the barrier was taken to be finite, have been reported after that by Masselink *et al.*[2,3] In a recent study, Pasquarello, Andreani, and Buczko also calculated the binding energies of the excited acceptor states.[4]

One of the recombination processes studied in this paper is usually referred to as a two-hole transition (THT).[5] This recombination process gives rise to THT peaks at a constant energy separation from the recombining bound exciton (BE), irrespective of the excitation energy. While such THT peaks are commonly observed in bulk material[5] the first report on a THT in QW's was not published until recently.[6] In addition to the observation of the $1s \rightarrow 2s$ transitions via THT's in photoluminescence (PL), the same transition was observed in resonant Raman scattering (RRS) for the Be acceptor in a QW for QW widths in the range 50–138 Å.[6,7]

SAMPLE PREPARATION AND EXPERIMENTAL CONDITIONS

The samples used in this study were grown by molecular-beam epitaxy (MBE) in a modular Varian Gen II system. The layers were grown on top of a semi-insulating (100) GaAs substrate having a 0.35-μm undoped GaAs buffer layer. The growth temperature was nominally 680 °C. The samples were grown under As_4-rich conditions and without interruptions at the QW interfaces. Five δ-doped samples were used in this study, all with the QW width of $L_z = 96$ Å ± 2 Å, but with different position of the dopant layers in the well, and one sample that was doped in the entire QW (sample *F*) with the QW width $L_z = 101$ Å (Table I). The final estimate of the QW widths was based on the energy positions of the free-exciton (FE) peaks in the PL spectra. The $Al_xGa_{1-x}As$ barriers were in all cases 150 Å wide and had an intended Al composition of $x = 0.30$, but a variation of the order $\Delta x = \pm 0.02$ between different growth runs was determined by measuring the near-band-gap PL peak position of the $Al_xGa_{1-x}As$ layers. All samples in-

TABLE I. Doping conditions: average deviation in dopant position from the center of the well, z_0; δ-dopant layer thickness, Δz; and concentration of Be acceptors, p.

Sample	z_0 (Å)	Δz (Å)	p (10^{17} cm^{-3})
A	0	20	1
B	13	6	1
C	27	6	1
D	37	6	2
E	45	6	1
F		101	2

vestigated contained multiple QW structures with 50 periods of alternating layers of GaAs and $Al_xGa_{1-x}As$. The samples were δ doped with Be at different positions between the center and the edge of the well (except for sample *F*, which was doped in the entire QW). The doping conditions are given in Table I.

For the PL measurements a Kr^+-ion laser ($\lambda=6471$ Å) was used as an excitation source. For the selective PL (SPL) measurements, a Kr^+-ion laser was used to pump a dye laser with a LD 700 dye. The laser beam was focused on the samples to a diameter of the order of 1 mm. The emitted light from the samples, perpendicular to the incident beam, was focused on the slits of a Jobin Yvon HRD1 0.60 m double-grating monochromator. All measurements presented in this study were performed at 2 K. The PL signal from the monochromator was detected with a dry-ice-cooled GaAs photomultiplier.

EXPERIMENTAL RESULTS AND DISCUSSION

The PL spectra shown in Fig. 1 represent the two extreme cases in this study. Figure 1(a) shows a PL spectrum for the center-doped sample (sample *A*). In addition to the FE and BE, the free electron to a hole bound at a neutral acceptor (FB) emission is also observed.

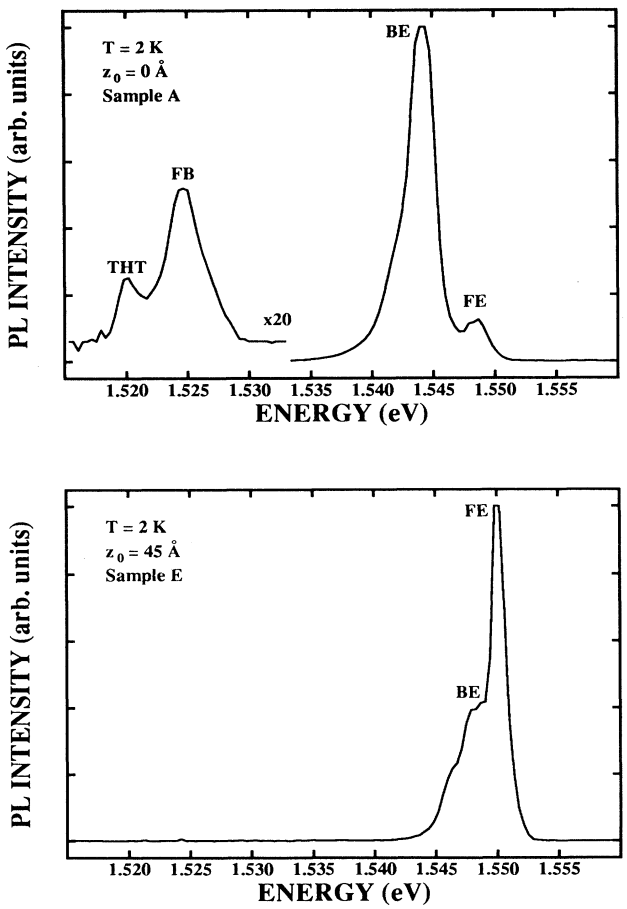

FIG. 1. PL spectra for (a) sample *A* ($z_0=0$ Å) and (b) sample *E* ($z_0=45$ Å) using a Kr^+ ion laser as an excitation source.

Even the THT is possible to see even though the excitation is nonresonant. This interpretation is confirmed by selective PL (SPL) measurements and is consistent with earlier measurements.[7] In Fig. 1(b) we show a PL spectrum for the edge-doped sample (sample *E*), for which the BE has approached the FE. On the low-energy side of the BE there is also a shoulder that has its origin in a BE bound at the Be acceptor at the center of the well. These Be acceptors exist at the center of the well probably because of diffusion and surface segregation during growth.[8] This has also been seen earlier[9] in a study of an edge-doped QW, where there was no intentional doping in the center, but still there was a strong FB luminescence attributed to Be acceptors at the center of the well.

Upon comparison of the two spectra in Fig. 1, we can see that the BE dominates for the center-doped sample, but the opposite is true for the edge-doped sample, i.e., the FE is much stronger than the BE. This is partly due to the larger amount of dopant atoms in the center-doped sample, which has a dopant layer thickness of ≈20 Å, compared to the edge-doped sample, which has a dopant layer thickness of only ≈6 Å. This explains only part of the difference in intensity. The remaining difference is probably due to differences in transition probability between center positioned BE's and BE's at the QW interface.

The binding energy of the BE, derived from the energy separation between the FE and BE, is shown in Fig. 2 as a function of the acceptor position in the well. The binding energy decreases, as expected, when the acceptors are displaced from the center of the well.

Figure 3 shows a synopsis of SPL spectra of sample *B* ($z_0=13$ Å). These spectra are typical for an off-center doped sample, because it is possible to detect THT and RRS from $1s\rightarrow 2s$ transition of acceptors at the main dopant position, denoted P_1 and R_1 respectively, as well as acceptors at the center of the well, denoted P_2 and R_2, respectively. When the excitation is resonant with or close to the light-hole (lh) or heavy-hole (hh) states of the FE, as observed in PL excitation (PLE), the THT's are enhanced. Similarly, the RRS transitions are enhanced

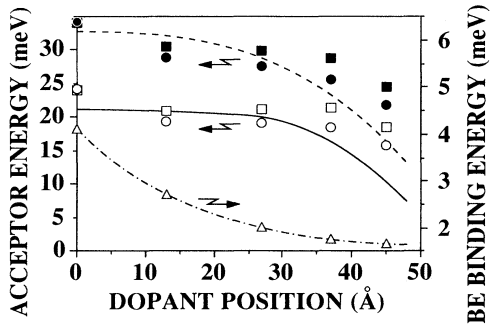

FIG. 2. BE binding energy, represented by open triangles vs the dopant position z_0 in the well for the samples used in the study. The dash-dotted line is a fit to the experimental data. The upper set of data corresponds to the $1s\rightarrow 2s$ acceptor transition energies and the total acceptor binding energies vs the dopant position z_0 in the well. The squares are from THT data and the circles are from RRS data.

when the excitation is resonant with or close to the BE peak in PL.[6] When the excitation energy is shifted towards the BE ($z_0=13$ Å) energy (1.5475 eV), the RRS of the Be acceptors at the dopant position, z_0, (peak R_1) is enhanced. When the excitation energy is shifted further and becomes below the BE ($z_0=13$ Å) energy, the intensity of peak R_1 decreases, and eventually disappears, while the intensity of peak R_2 increases and has a maximum at the energy corresponding to the energy of the BE ($z_0=0$ Å). Shifting the excitation energy further towards lower energy, below the BE ($z_0=0$ Å) bound at a center positioned acceptor, the intensity of peak R_2 decreases and eventually disappears. The energy separation between the excitation energy and peak R_2 is constant, which is characteristic for RRS peaks. Worthwhile to notice is that the energy displacement between the excitation and peak R_1, corresponding to the $1s\rightarrow 2s$ transition, is not constant but increases with decreasing excitation energy, i.e., with increasing BE binding energy within the width of the BE peak. The increased energy displacement has been found to be due to spatially selective excitation of the acceptors both within and outside the δ-dopant layer.[10] This fact is due to the distribution of acceptor binding energies within the finite δ-dopant layer, which gives rise to a corresponding distribution of BE binding energies.

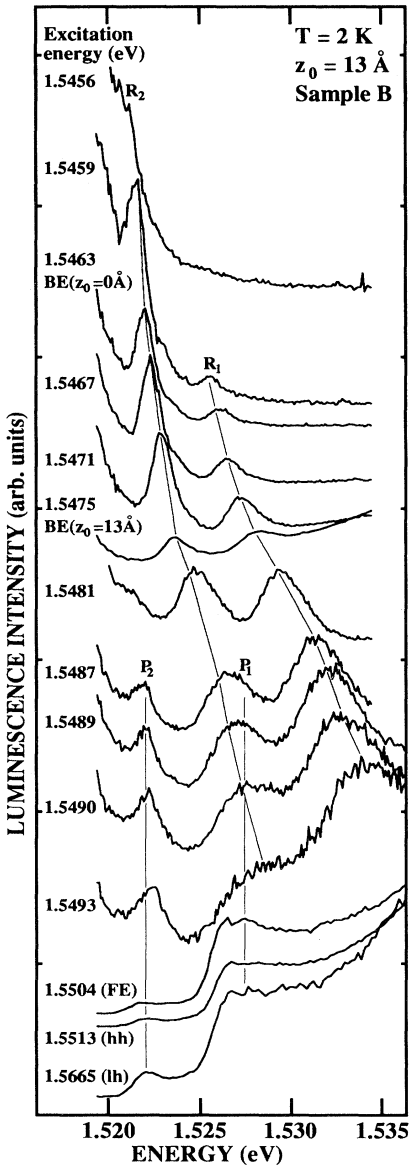

FIG. 3. SPL spectra of sample B ($z_0=13$ Å) using an excitation intensity of ≈ 100 mW/mm^2. The notations (hh) and (lh) denote the heavy-hole and light-hole states of the FE as observed in PLE, and (FE), [BE ($z_0=13$ Å)], and [BE ($z_0=0$ Å)] denote the FE and the BE, bound to acceptors at $z_0=13$ Å and at the center of the well ($z_0=0$ Å), respectively, as observed in PL.

Why do the center-related peaks P_2 and R_2 appear in the spectra of the off-center doped QW's (Fig. 3)? Due to the displacement of the acceptor hole wave function for an off-center positioned acceptor, the overlap between the acceptor hole-wave function and the BE wave function decreases. This hole wave function displacement has its maximum ≈ 38 Å from the edge for an acceptor at the edge of a 100-Å-wide QW.[2] This will give rise to a significant overlap for acceptors and BE's at the center of the well also for off-center doped samples. The intensity of the center acceptor THT's is enhanced by the density of states per energy, $g_L(E)=(2/L)(dz_0/dE)$, which increases towards the center and becomes infinite at the center since dE/dz_0 vanishes at $z_0=0$.[1,2]

The $1s\rightarrow 2s$ transition energy of the Be acceptor in the center-doped QW is, in this work, found to be 24.1 ± 0.5 meV from THT data and 24.3 ± 0.5 meV from RRS data, which agrees well with recent measurements.[7] Comparing this with the values for a center positioned acceptor in the off-center doped samples, we find that all the measured $1s\rightarrow 2s$ acceptor transition energies for the different samples (samples A–E) lie within a 1-meV-wide interval. This supports the above interpretation of the peaks P_2 and R_2 in Fig. 3.

The results from the THT and RRS measurements are summarized in Fig. 2 for the different dopant positions, represented by the open symbols. The solid line in the same figure is the theoretical prediction for the $1s\rightarrow 2s$ transition energy of a Be acceptor by Pasquarello, Andreani, and Buczko.[4] It should be pointed out that the theoretical calculations[4] have been performed for a slightly higher x value, $x=0.4$. The dashed line is the theoretical prediction for the total binding energy of a Be acceptor by Masselink *et al.*[2] The solid symbols represent the experimentally determined total binding energies, where the $1s\rightarrow 2s$ part is from our measurements to which the binding energies of the $2s$ states from the calculations by Pasquarello, Andreani, and Buczko[4] is added.

Measurements of a QW that is homogeneously doped in the entire QW (sample F) show clearly the FB luminescence from acceptors at the center of the well as well as the FB luminescence from acceptors at the edge. The total binding energies, estimated from these transitions, is 34 ± 1 meV for acceptors at the center of the well

and 18±1 meV for acceptors at the QW edge. Upon comparison of these values with the experimental data for QW's of widths in the range 78–149 Å of Masselink *et al.* (Fig. 8 in Ref. 2), it is obvious that a total binding energy of roughly 18 meV for acceptors at the edge is expected, whereas our value, 34 meV (Fig. 2), for acceptors at the center of the well is somewhat higher than the 32 meV that they estimated from FB transitions. The total binding energy of acceptors at the center of the well can also be estimated from the FB transition of a center-doped QW shown in Fig. 1(a). This estimate gives a total binding energy of roughly 34 meV, consistent with the other estimates. A $1s \rightarrow 2s$ acceptor transition energy of 15.8 meV is derived for the dopant layer position in the edge-doped QW determined from the RRS results and are slightly higher for the THT measurements. Our experimental results on the total binding energies and $1s \rightarrow 2s$ acceptor transition energies at the edge disagree with theoretical prediction but agree well with earlier experimental estimates. The latter statement is based on the fact that the spatial separation between the interface and the dopant layer center corresponds to roughly 2.5 meV. It should be noted that the theoretical calculations[4] correspond to a higher x value, $x=0.4$, than for the samples used ($x=0.3$). A lower x value will actually increase the binding energy for an acceptor at the edge,[2] while the effect on an acceptor in the center of the QW is negligible. This fact will thus reduce the disagreement between the theoretical predictions and our results shown in Fig. 2. To conclude, the provided binding energies and $1s \rightarrow 2s$ acceptor transition energies are consistent with earlier experiments as well as theory within a few meV for the center-doped QW's, while our data at the edge disagree to some extent with the theory, illustrated by Fig. 2, but are consistent with earlier experimental results.

If an acceptor is introduced into the center of a QW instead of bulk, the symmetry is reduced from point T_d group to D_{2d}. For an off-center position ($z_0 \neq 0$) the symmetry is further reduced to C_{2v}.[4] The consequence of this is that the fourfold degenerate Γ_8 band in the T_d symmetry splits into two twofold degenerate bands Γ_6 and Γ_7 in the D_{2d} symmetry.[2,3] Of these states (Γ_6 and Γ_7), Γ_6 has the lowest energy. At low temperatures the main occupation will be in this lower state and at $T=2$ K almost all of the measured $1s \rightarrow 2s$ acceptor transitions will be in the Γ_6 symmetry. The selection rules, which apply for RRS as well as THT in the case of acceptors in bulk, strongly favor transitions between states of the same parity, i.e., mainly from $1s_{3/2}$ ground state to s-like excited states.[11] Accordingly, for the case of impurities confined in a QW, the strongest line observed in RRS is ascribed to $1s \rightarrow 2s$ transitions, for donors[12] as well as for acceptors.[13] For the case of C_{2v} symmetry ($z_0 \neq 0$), all eigenstates transform like Γ_5. The observed transitions in THT and RRS are then attributed to the $1s(\Gamma_5) \rightarrow 2s(\Gamma_5)$ transitions.

Based on the facts stated above, the interpretation of the peaks P_1 and R_1 observed in THT and RRS, respectively, is the $1s_{3/2} \rightarrow 2s_{3/2}$ transition of the Be acceptor at position z_0. The peaks P_2 and R_2 are thus interpreted as the $1s_{3/2} \rightarrow 2s_{3/2}$ transition of a Be acceptor at the center of the well. There is a reasonably good agreement between the transition energies in THT and RRS for all the samples. The observed discrepancy between THT's and RRS, especially for the edge-doped QW, can be explained by the displacement of the acceptor hole wave function mentioned above. This displacement will give a THT energy corresponding to acceptors closer to the center of the well than expected from the position of the δ-dopant layer; the size of the energy difference between the THT's and RRS will depend on the actual profile of the δ-dopant layer. As can be seen in Fig. 2, the difference between the THT's and the RRS decreases towards the center of the well, which can be expected since the z variation of the $1s \rightarrow 2s$ acceptor transition energy is reduced with decreasing z_0.

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