Evidence for resonant electron capture and charge buildup in GaAs/Al_xGa_{1-x}As quantum wells

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Evidence for the resonant capture of electrons into quantum wells (QW's) is demonstrated in the photoluminescence (PL) of GaAs/Al_xGa_{1-x}As QW's doped with Be. We find that the PL intensity ratio between the free-to-bound and excitonic transitions exhibits a strong oscillation as a function of the QW width bearing a striking resemblance to the theoretical prediction of the electron capture rate. It is shown that this PL behavior reflects the buildup of excess negative charge in the QW arising from the different capture efficiencies of electrons and holes. [S0163-1829(96)04623-1]

Capture of carriers into quantum wells (QW's) has been studied with considerable interest aimed at optimizing carrier collection efficiency for improved performance of QW lasers. Of particular interest is the quantum mechanical resonance in the capture of carriers. Brum and Bastard[1] and Kozyrev and Shik[2] have predicted a strong oscillation of the capture time as a function of the well width originating from two different resonance mechanisms. The first kind of resonance is associated with a so-called virtual bound state, which appears in the barrier continuum when the highest QW level lines up with the band edge of the barrier. In this situation, the barrier wave function penetrates largely into the QW region, thus increasing the overlap with the QW bound states and enhancing the capture rate. The second kind of resonance occurs when one of the QW levels lies just one LO-phonon energy below the barrier band edge. In this case, the in-plane momentum of the emitted phonon becomes zero. Since the matrix element of electron-LO-phonon interaction varies as $1/\mathbf{q}^2$, where $\hbar\mathbf{q}$ is the momentum of phonon, this also leads to the enhancement of the capture rate. Experimentally, several groups have successfully demonstrated the resonant capture of electrons with the help of subpicosecond luminescence techniques[3–5] and/or samples with special structures.[3,4,6] These studies have shown that such resonance effects do exist and can be observed in appropriate conditions.

In this paper, we present photoluminescence (PL) studies of Be-doped QW's, which provide evidence for the resonant capture of electrons. We find that the intensity ratio between the free-to-bound and excitonic transitions exhibits a strong oscillation as a function of the well width. It is shown that this PL behavior reflects the buildup of negative charge in the QW arising from the different capture efficiencies of electrons and holes.

The samples studied are GaAs/Al_xGa_{1-x}As single QW structures grown at 580 °C on semi-insulating GaAs(100) substrates by molecular-beam epitaxy. The GaAs QW was δ doped at the well center with [Be]$=10^{10}$ cm^{-2}. The Al_xGa_{1-x}As barriers were 500 Å thick and the Al composition was varied as $x=0.22$, 0.26, and 0.31, as determined from the band gap of the Al_xGa_{1-x}As barriers. QW's with various well widths and the same x were prepared in one wafer by suspending the substrate rotation during the growth of the GaAs well, thereby allowing the well width to vary across the wafer as a result of the spatial nonuniformity of the Ga beam flux.[7] PL was measured at 7 K with an Ar^+-ion laser as the excitation light source unless specified. Typical excitation power was 50 μW, which corresponds to a power density of $\approx$0.3 W cm^{-2}.

Figures 1(a) and 1(b) show the PL spectra of $x=0.31$ QW's with various well widths L_z. The peaks labeled FE and BE are due to the free excitons and excitons bound to neutral acceptors, respectively. The peaks labeled (e,A^0) are due to free-to-bound transition, in which electrons in the first

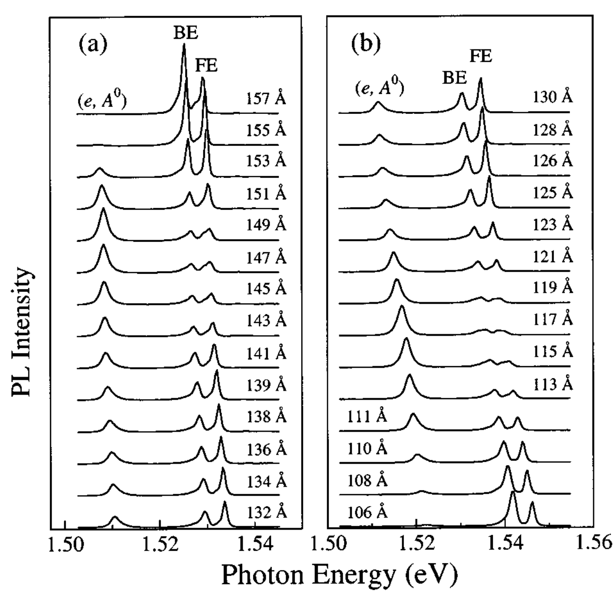

FIG. 1. PL spectra of $x = 0.31$ QW's with various well widths. The well widths were calculated from the energy positions of the FE peaks.

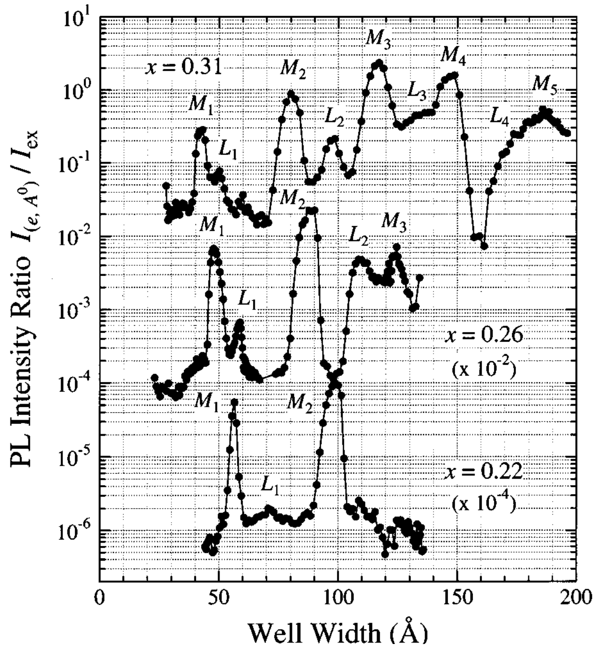

FIG. 2. PL intensity ratio between the free-to-bound and excitonic (FE plus BE) transitions as a function of the well width. For clarity, the results for $x=0.22$ and 0.26 are shifted vertically.

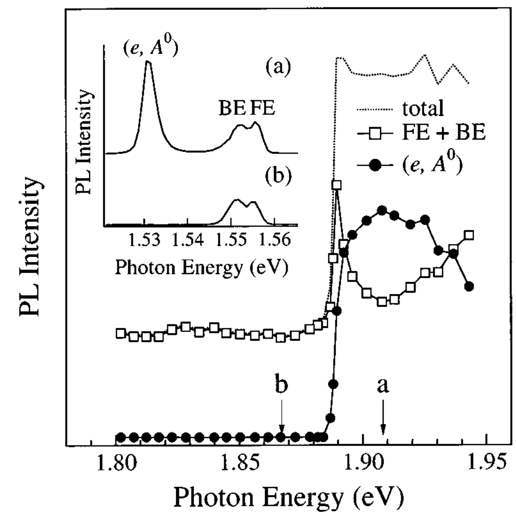

FIG. 3. Excitation-energy dependence of the PL intensities of free-to-bound and excitonic transitions for 90-Å QW with $x=0.26$. Inset: PL spectra for the excitation (a) above and (b) below the barrier band gap. The excitation energies are indicated by the arrows.

conduction subband recombine with holes bound to Be acceptors. It is seen that these peaks exhibit remarkable intensity variations with L_z. It is noteworthy that the behavior of the free-to-bound and excitonic transitions are complementary. As the free-to-bound transition develops, the excitonic peaks shrink, and vice versa, keeping the total PL intensity almost constant.

The PL intensity ratio between the free-to-bound and excitonic transitions is plotted in Fig. 2 as a function of the well width, together with the results for $x=0.22$ and 0.26. The figure reveals a strong oscillation of the PL intensity ratio. For $x=0.31$, up to five pronounced peaks labeled M_n ($n=1$–5) appear in series, accompanied by subsidiary peaks labeled L_n ($n=1$–4). As x is decreased, the oscillation period gets longer and the peaks shift toward larger well widths. Apparently, these oscillations bear a striking resemblance to the oscillatory behavior of the carrier capture rate predicted in Refs. 1 and 2.

The importance of carrier capture in the above PL behavior is confirmed by changing the excitation photon energy below and above the barrier band gap. Figure 3 depicts the excitation-photon-energy dependence of the integrated intensities of the free-to-bound and excitonic transitions for a 90-Å QW with $x=0.26$. The PL spectra obtained for the excitation above and below the barrier band gap ($=1.89$ eV) are compared in the inset. Recall that in Fig. 2 the intensity of the free-to-bound transition peaks at this well width ($L_z=90$ Å for $x=0.26$). Figure 3 shows that such an enhancement of the free-to-bound transition occurs only for the excitation *above* the barrier band gap. For the excitation *below* the barrier band gap, the free-to-bound transition is absent and, accordingly, the intensity ratio oscillation was not observed. This result confirms that the carrier transfer from the barrier into the QW plays an essential role.

In Fig. 4, the peak positions of the PL intensity ratio are compared with the well widths for which the resonant capture of electrons is predicted. In calculating the position of the resonance, the nonparabolicity of the conduction band[8] was necessarily taken into account, and the conduction-band offset ratio was assumed to be 0.62.[9] As seen in the figure, the positions of M_n and L_n peaks are coincident with the predicted positions of the first and second kinds of resonances (hereafter referred to as virtual-bound-state and LO-phonon resonances), respectively. From the overall agreement in the peak positions and their dependence on x, M_n

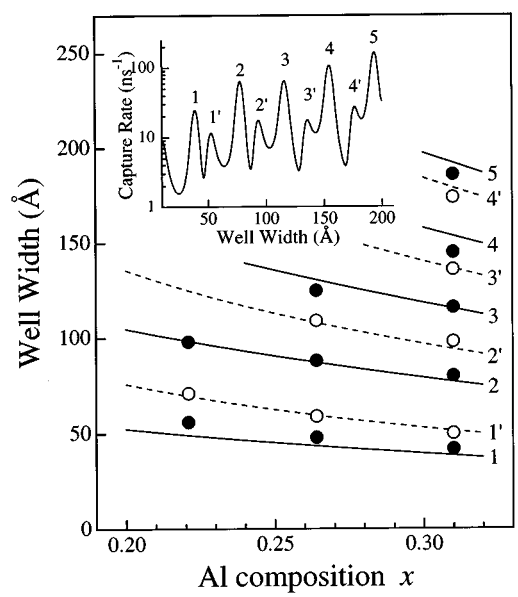

FIG. 4. Comparison of the peak positions of the PL intensity ratio and the resonance in electron capture. M_n and L_n peaks are plotted as closed and open circles. The virtual-bound-state and LO-phonon resonances are shown as solid and dashed lines labeled n and n', respectively. Inset: calculated electron capture rate vs well width.

and L_n peaks are assigned to the virtual-bound-state and LO-phonon resonances, respectively.[10]

This assignment is also consistent with the amplitude of the resonance peaks. The inset of Fig. 4 depicts the electron capture rate via LO-phonon emission calculated assuming the 7-K Boltzmann distribution of barrier electrons. As seen in the figure, the resonance peak becomes larger as n increases, and the virtual-bound-state resonances are consistently larger than the LO-phonon resonances.[11] These tendencies are consistent with the amplitude of M_n and L_n peaks, supporting our assignment.

Now we discuss the mechanism responsible for the observed PL behavior. Let N^0, N^-, n, and p be the densities of neutral and ionized acceptors, free electrons, and free holes in the QW, respectively. Noting that the recombination rate of excitons should be equal to their formation rate in the steady state, the PL intensities of the free-to-bound and excitonic transitions are given as $I_{(e,A^0)}=AN^0n$ and $I_{\text{ex}}=Bnp$, respectively, where A and B are constants. The acceptors are ionized by the free-to-bound transition, which are then neutralized by getting free holes. This detailed balance requires that $AN^0n=CN^-p$, where C is a constant. Thus, the PL intensity ratio is given by

$$\frac{I_{(e,A^0)}}{I_{\text{ex}}}=\frac{A}{B}\frac{N^0}{p}=\frac{C}{B}\frac{N^-}{n}. \tag{1}$$

Using the total acceptor density N $(=N^0+N^-)$, Eq. (1) can be rewritten as

$$\frac{I_{(e,A^0)}}{I_{\text{ex}}}=\frac{N/B}{p/A+n/C}. \tag{2}$$

Since A, B, C, and N in Eq. (2) are constants, the oscillation of the PL intensity ratio implies that n and p should oscillate with the well width. It is important to note that, for a fixed carrier generation rate, n and p can be varied only by breaking the charge neutrality within the QW, i.e., $N^-+n-p\neq 0$.

It is therefore suggested that the role of the resonant electron capture is to break the charge neutrality in the QW through preferential capture of electrons. The accumulation of negative charge in the QW results in the band bending, which in turn modulates the capture rates of electrons and holes; the hole capture rate is enhanced and the electron capture rate is reduced until they become equal in the steady state. Hence, the charge density in the steady state should reflect the difference between the initial capture efficiencies of electrons and holes, and thus provides a measure of the capture efficiency of electrons.[12]

The effect of the charge imbalance on the carrier recombination in doped QW's is demonstrated by the following rate-equation analysis. As discussed above, in the steady state electrons and holes are injected into the QW at the same rate G. Thus, the rate equations are written as $G-Bnp-AN^0n=0$ for electrons and $G-Bnp-CN^-p=0$ for holes. These coupled equations have been solved numerically to obtain n, p, N^0, and N^- as functions of the excess charge density $\delta\equiv N^-+n-p$.

The results of the calculation are displayed in Fig. 5(a). In the calculation, we assumed that $G=3\times10^{16}$ $\text{cm}^{-2}\,\text{s}^{-1}$,

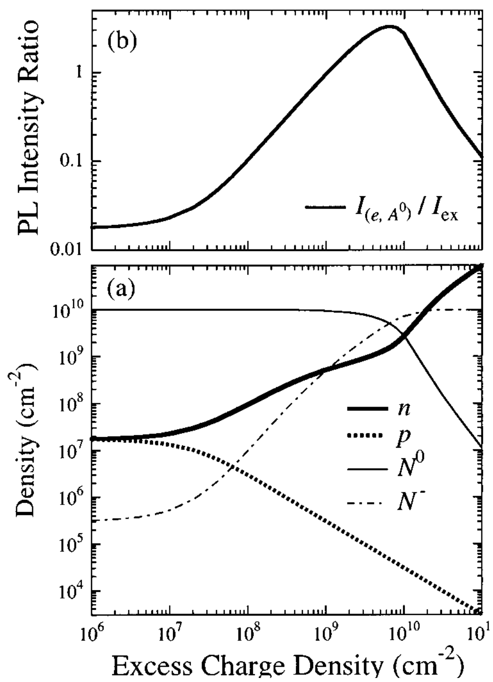

FIG. 5. (a) Densities of electrons, holes, and neutral and ionized acceptors and (b) PL intensity ratio between the free-to-bound and excitonic transitions, calculated as functions of the excess charge density in the QW.

$A=3\times10^{-3}$ $\text{cm}^{-2}\,\text{s}^{-1}$, and $B=C=10^2$ $\text{cm}^{-2}\,\text{s}^{-1}$. These parameters were chosen by fitting the results of time-resolved PL experiments, which will be published separately. The calculation demonstrates that n, p, N^0, and N^- are strongly dependent on δ. This implies that the addition of extra electrons to the QW significantly affects the population of holes and the charge state of the acceptors.

As a result, the PL intensity ratio between the free-to-bound and excitonic transitions changes significantly with the excess charge density, or the electron capture efficiency, as shown in Fig. 5(b). The intensity ratio changes by more than two orders of magnitude, from ~0.02 to ~3, as δ is increased from 10^6 to $\sim7\times10^9$ cm^{-2}. The observed variation of the PL intensity ratio has thus been reproduced by assuming the buildup of excess negative charge in the QW.

The above PL behavior is understood as follows. When the electron and hole capture efficiencies are nearly equal and δ is smaller than 10^7 cm^{-2}, most of the acceptors remain neutral as seen in Fig. 5(a). Holes can therefore form excitons without being captured by ionized acceptors. Consequently, the excitonic recombination is dominant in this case. As the electron capture efficiency is enhanced and δ exceeds 10^7 cm^{-2}, p decreases rapidly. This is because the free holes are used up in the exciton formation with the excess electrons. Since these excess electrons are left unable to find free holes, they recombine with holes bound to acceptors. This explains the striking enhancement of the free-to-bound transition and the quenching of the excitonic transitions in the resonant condition. This can also be understood by Eq. (1), in which p decreases with increasing δ while N^0 remains almost constant.

It is seen that the PL intensity ratio declines for the further increase of δ above 10^{10} cm^{-2}. As understood from Eq. (1), this is because N^- saturates at 10^{10} cm^{-2} while n continues to increase.[13] For the present case, however, the excess charge density is thought to be less than 10^{10} cm^{-2}, since the PL intensity ratio depends monotonically on the electron capture rate.[14]

Finally, we compare our results with previous reports on undoped QW's. The present study shows that strong resonance effects are operating even in steady-state PL of single QW's, in contradiction to some pervious studies showing no evidence for the resonance effects. This is understood as follows. Even if excess charge builds up in intrinsic QW's, it influences the PL only a little, since all the carriers finally recombine as free excitons.[15] The role of Be acceptors in our study is to provide a competing recombination channel for the excess electrons. This enables us to observe the charge buildup and the resonant capture of electrons.

In summary, we have demonstrated evidence for the resonant capture of electrons in PL of Be-doped QW's. The observed oscillation of the PL intensity ratio between the free-to-bound and excitonic transitions has been explained as being due to negative charge buildup in the QW arising from the different capture efficiencies of electrons and holes. Although this phenomenon is observed only in doped QW's, our results indicate that the resonant capture and the charge buildup can occur also in intrinsic QW's.

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[7] For $x=0.31$, we have grown two wafers to cover the wide range of well widths.

[8] T. Ruf and M. Cardona, Phys. Rev. B **41**, 10 747 (1990).

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[10] We have also calculated the positions of the hole capture resonances to find that neither heavy- nor light-hole resonances can explain the experimental observations.

[11] The relative contribution of the two resonance mechanisms is opposite in Ref. 1 This is because the authors of Ref. 1 assumed that the initial barrier states were equally populated within 36 meV from the conduction-band bottom of the barrier. We assumed the Boltzmann distribution as in Ref. 2, which we believe to be more realistic.

[12] The resonant capture of holes has not been reported so far. We therefore assume that the capture efficiency of holes does not depend on the well width so significantly as that of electrons.

[13] We find that the maximum of the PL intensity ratio always occurs for $N^0=N^-$, and the δ value for the maximum does not significantly change with the carrier generation rate.

[14] For $\delta=10^{10}$ cm^{-2}, the induced electric field is 1.4×10^3 V cm^{-1}, corresponding to 7-meV potential variation across the 500-Å $Al_xGa_{1-x}As$ barrier. This may be enough to equalize the electron and hole capture rates.

[15] This is not the case in the presence of nonradiative centers. [A. Fujiwara *et al.*, Phys. Rev. B **51**, 14 324 (1995).]