

Nonuniform exciton localization in different GaAs quantum wells studied by spatially resolved cross-sectional cathodoluminescence

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Received zzz, revised zzz, accepted zzz

Published online zzz (Dates will be provided by the publisher.)

PACS 78.60.Hk, 78.67.De, 73.63.Hs, 73.50.Gr

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Excitonic radiative recombination in different GaAs quantum wells (QWs) is investigated by cross-sectional cathodoluminescence (CL) spectroscopy with high spatial resolution. By measuring spectrally discriminated CL intensities originating from the various QWs as a function of excitation position, the exciton bands of the indi-

vidual QWs can be mapped to see how the respective CL intensities are influenced by the excitation position along the growth direction. We find that nonuniform capture of nonequilibrium electron-hole pairs by the QWs plays an important role for the exciton recombination, which strongly depends on the traversing pathway.

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1 Introduction The luminescence intensity and dynamics from semiconductor quantum wells (QWs) embedded in heterostructures are in most cases assumed to originate from uniformly populated electron-hole pairs in the wells after the initial rapid relaxation of carriers from the barriers, where they are generated. However, when there are several different wells or in inhomogeneous QW systems, the luminescence intensity should be influenced by the pathway during the relaxation except for resonant capture processes [1-3]. This is especially true for the QW systems with vertical (along the growth direction) and lateral potential fluctuations. In this paper, excitonic radiative recombination is investigated in GaAs QWs differing in their thickness embedded in a separate-confinement heterostructure (SCH) by cross-sectional cathodoluminescence (CL) spectroscopy. By measuring spectrally discriminated CL intensities originating from the various QWs as a function of excitation position, we are able to spatially resolve the exciton bands of the individual QWs. We find that the CL intensities of the various exciton bands strongly depend on whether there exist carrier-trapping sites between the excitation and recombination positions or not.

2 Experimental The sample used for this study consists of three different QWs grown on an undoped GaAs (100) substrate by molecular beam epitaxy, prepared with growth interruption at the QW interfaces [4-6]. The nominal widths of the GaAs QWs, which are separated from each other by 36 nm thick $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ inner barriers, amount to 7.8 nm (QW1), 5.5 nm (QW2), and 3.5 nm (QW3) starting from the substrate side. The three QWs and the two inner barriers are embedded in a pair of 72 nm $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ barriers. This whole structure is additionally confined by 0.19 μm (surface side) and 0.43 μm (substrate side) $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ outer barriers, forming a SCH configuration to avoid surface nonradiative recombination. These structures were grown nominally undoped with an estimated background doping level due to carbon below 10^{14} cm^{-3} . Depth-resolved cross-sectional CL spectroscopy was performed on the cleaved edge of the sample in a scanning electron microscope at 6 K, using an electron beam energy of 3 keV and a beam current of 0.1 nA. A spectrometer with a focal length of 0.3 m and a cooled CCD array detector were used to disperse and detect the CL signal, respectively. The spatial resolution of the CL measurement amounts to about 50 nm.

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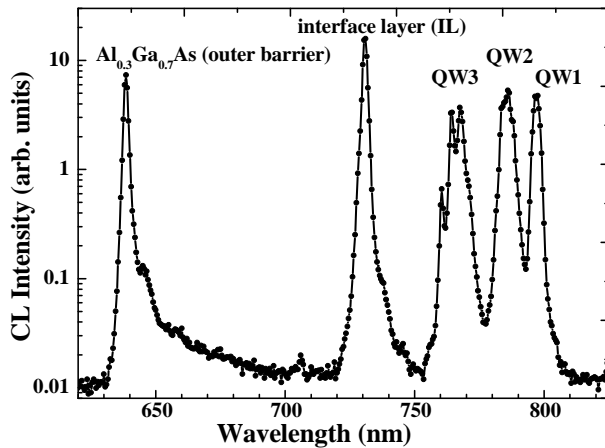


Figure 1 CL spectrum for excitation normal to the sample surface at 6 K. The CL intensity is plotted on a logarithmic-scale. Five emission bands are observed due to QW1, QW2, QW3, the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barrier layer, and an interface layer (IL) formed between the bottom $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ and $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers.

3 Results and Discussion Figure 1 shows a typical CL spectrum of the sample, with the CL intensity plotted on a logarithmic scale, which was taken normal to the growth surface. In the wavelength range between 750 and 810 nm, three exciton emission bands due to QW1, QW2, and QW3 are observed in agreement with our previous studies [7, 8]. Note that the spectral fine structure due to the formation of monolayer growth islands is clearly seen for the narrowest QW3 because of the largest energy splitting in exciton lines (~ 7 meV) by one monolayer change of the well thickness. A CL peak observed at 640 nm is ascribed to the exciton recombination in the outer $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers. A strong peak observed around 728 nm is attributed to an unintentionally introduced interface layer (IL) formed between the bottom $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ and $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers due to the Al cell temperature overshoot (when the Al cell was set at a lower effusion cell temperature corresponding to the $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ layer). The presence of this IL is confirmed by cross-sectional transmission electron microscopy (TEM). Figure 2(a) displays a dark-field TEM image obtained with the diffraction vector $\mathbf{g}=002$, which is sensitive to the chemical composition of the alloy. As a consequence, the GaAs QWs appear in dark contrast, while the (Al,Ga)As inner and outer barriers show different grey contrasts depending on the Al content. The presence of the IL is clearly observed. Figure 2(b) shows an intensity profile after a line-scan on the marked area, where the presence of the IL is clearly seen.

In Fig. 1, a faint CL peak centred at 706 nm appears, which is attributed to the $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ inner barrier layer. It is very small because carriers generated within the barriers are efficiently trapped by the QWs. This assignment can be

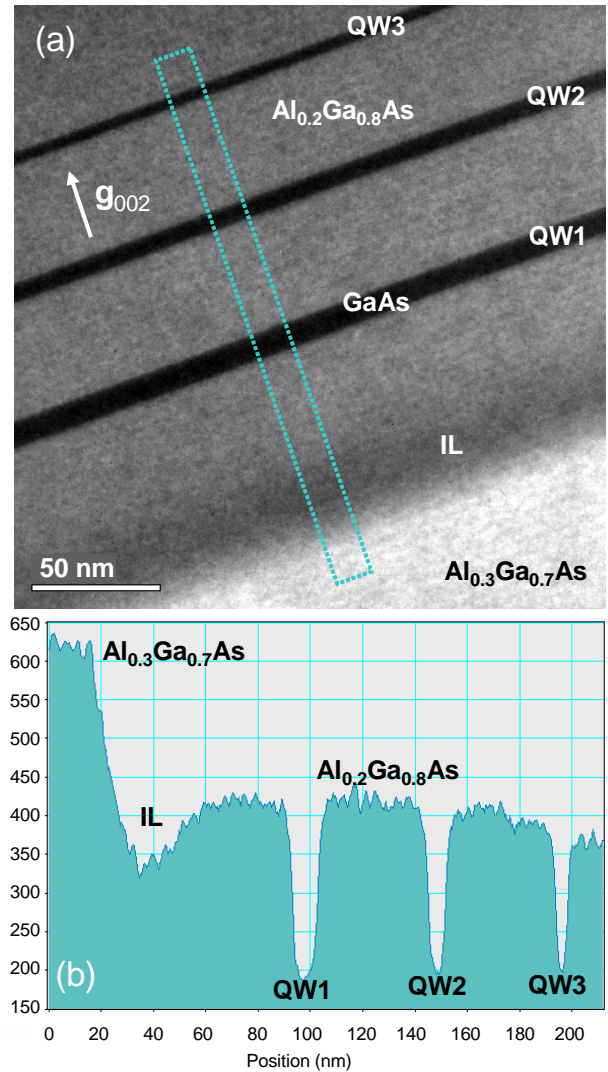


Figure 2 (a) Cross-sectional TEM image obtained with the chemically sensitive $\mathbf{g}=002$ diffraction vector. (b) Intensity profile obtained after an average line-scan on the marked area in (a) clearly showing the presence of the IL.

confirmed by measurements of photoluminescence excitation spectra [8].

A series of spot CL spectra for varying excitation positions (with a step length of $0.017 \mu\text{m}$) were recorded by moving the electron-beam position across the heterostructure on the cleaved edge of the sample (CL line-scan). Figure 3 shows the integrated CL intensities of the four emission bands (QW1, QW2, QW3, and IL) at 6 K as a function of electron-beam position along the growth direction. The CL intensities of the $n = 1$ exciton states of QW3, QW2, and QW1 exhibit successive maxima as the scan position moves towards the substrate exactly in order of the growth sequence and at the expected position. In Fig. 3, QW3 shows the highest CL peak intensity among the four exciton bands. We attribute it to the strongest confinement

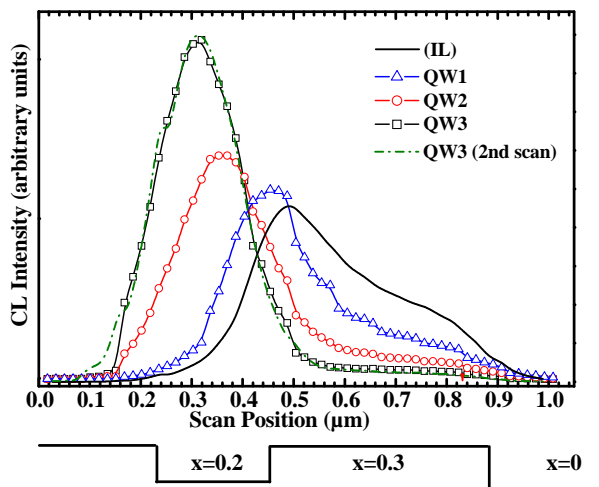


Figure 3 Cross-sectional CL intensity distributions measured on the cleaved edge of the sample for four bands (QW1, QW2, QW3, and IL) at 6 K.

and to the efficient carrier sink due to resonant capture [7].

On the surface side of the $0.19 \mu\text{m}$ $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ outer barrier (scan position at $0.0\text{-}0.2 \mu\text{m}$), the CL intensities of the four bands are small due to surface nonradiative recombination, while they show characteristic and systematic differences on the substrate side of the $0.43 \mu\text{m}$ outer barrier (scan position at $0.5\text{-}0.8 \mu\text{m}$). Note that, when the electron beam excites the bottom barrier center (e.g., for a position at $0.65 \mu\text{m}$), the CL intensity for QW3 is smallest and the one for QW2 is smaller than that for QW1. This means that the generated carriers have difficulty to reach the QW3 layer due to intervening trapping sites of QW1 and QW2 during the diffusion processes. If the beam position is located further into the GaAs buffer layer ($> 0.9 \mu\text{m}$), the probability for the generated carrier pairs captured by the four bands is strongly reduced because of the existence of the GaAs carrier sink with a low band gap energy.

In order to examine the detailed distribution of the CL intensity as a function of excitation position, the logarithm of the CL intensities of the four bands is plotted in Fig. 4. Since the CCD detector does not allow to acquire the complete spectral range in one shot, two line scans along the same pathway were performed. For the first and second one, the spectra of the three QWs and the spectra of QW3 as well as IL have been acquired, respectively. Both profiles are then superimposed, where the CL intensity distribution of QW3 is used to calibrate both pathways. If we look at the bottom outer barrier region, the CL intensity of IL shows a single exponential decay as the excitation position goes away from the IL, indicating a diffusion-limited process operative for the ambipolar carrier transport from the excitation position towards the recombination site.

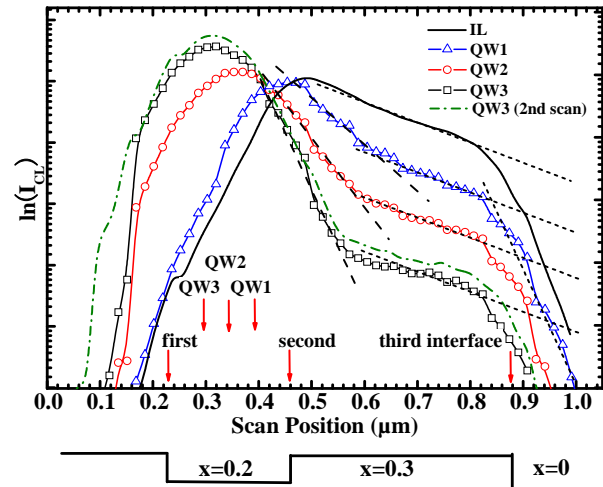


Figure 4 Cross-sectional CL intensity distributions plotted on a logarithmic scale for the four bands (QW1, QW2, QW3, and the interface layer) at 6 K. Dashed lines are guides to the eye.

Therefore, from this slope, we deduce a diffusion length $L = (D\tau)^{1/2}$ of $0.3 \mu\text{m}$ in agreement with the previous study [9]. In the outer barrier region far from the recombination sites ($0.6\text{-}0.8 \mu\text{m}$ region), the CL intensities of QW1, QW2, and QW3 show a similar diffusion-limited exponential slope. However, the absolute CL intensity is significantly decreased as the excitation position moves away from the recombination site because of the existence of carrier trapping sites. As the number of carrier trapping sites between the excitation and detection positions is increased, the absolute CL intensity decreases. The CL intensity of QW3 farthest from the bottom outer barrier thus shows the lowest value. When the excitation position is closer to the trapping sites, the diffusion-limited transport is significantly modified for scan positions between 0.3 and $0.55 \mu\text{m}$, for which the CL intensity distribution exhibits an increasing exponential slope for QW1 to QW3. That is, it becomes steepest for QW3 because of the additional shortening of the electron-hole pair lifetime by capture due to the existence of carrier traps of IL, QW1, and QW2. This change of the slope is easily understood in terms of the reduced lifetime of carriers by increasing the number of carrier trapping sites. If we look at the front outer barrier region ($0.1\text{-}0.4 \mu\text{m}$) in Fig. 4, a reversed tendency of the CL intensities is observed. That is, the CL intensity of IL is lowest and the slope is steepest. These results of the excitation position dependence of the CL intensity profiles indicate that the intensity of excitonic radiative recombination strongly depends on the traversing pathways and is not always uniform across the heterostructure.

4 Conclusion

In summary, cross-sectional CL inten-

1
2 sity distributions due to excitonic radiative recombination
3 are investigated in GaAs QWs differing in their thickness
4 in a separate-confinement heterostructure configuration.
5 By measuring spectrally discriminated CL intensities
6 originating from the various QWs versus excitation posi-
7 tion, we are able to spatially resolve the exciton bands of
8 the individual QWs and to see how the respective CL in-
9 tensities are influenced by the excitation position relative
10 to the recombination site. The CL intensity distribution
11 limited by diffusion transport is observed, when there are
12 no intervening trapping sites. However, the existence of
13 trapping sites between the excitation and recombination
14 sites significantly affects the CL intensity distributions by
15 the capture of generated carriers. We thus find that exciton
16 localization into QWs is not always uniform and strongly
17 depends on the traversing pathway in inhomogeneous QW
18 systems.
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