Influence of composition fluctuations in Al(Ga)As barriers on the exciton localization in thin GaAs quantum wells

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(Received 22 August 1996)

The localization of excitons in thin GaAs/Al(Ga)As quantum wells has been investigated by micro- and time-resolved photoluminescence (PL) spectroscopy. A fine-structured line shape in micro-PL is found not only for exciton recombination in the GaAs well but also, very similarly, for the Al(Ga)As barrier luminescence. By means of time-resolved measurements we show that the observed barrier luminescence probes selectively a barrier region close to the top interface of the quantum well. Our results directly reveal the essential contribution of Al(Ga)As composition fluctuations to the excition localization in GaAs/Al(Ga)As quantum wells. [S0163-1829(97)06208-5]

Optical and electronic properties of semiconductor heterostructures are influenced by their microscopic interface structures. Photoluminescence (PL) spectroscopy is a common tool used to characterize quantum-well (QW) structures in which the importance of the interfaces increases with decreasing QW thickness. Recently, the application of high spatial resolution optical techniques has revealed that the exciton lines in conventional PL spectra of thin GaAs/Al(Ga)As QWs in general consist of many narrow lines.[1–3] This fine structure can be explained by exciton localization at distinct minima of the random lateral potential landscape for the center-of-mass motion.[4,5] The potential fluctuations have commonly been discussed in terms of QW thickness fluctuations caused by interface roughness.[6] Composition fluctuations in the Al(Ga)As barriers were thereby often considered as playing a minor role; however, from the experimental and theoretical work done so far, it has not been possible to decide which is the main source for the exciton localization. In this work, we study micro- and time-resolved PL properties of thin state-of-the-art GaAs/Al(Ga)As QW structures and we demonstrate that composition fluctuations in the barriers influence substantially the QW potential fluctuations.

Three single QW (SQW) structures prepared by molecular-beam epitaxy on GaAs(001) substrates were studied. SQW 1 consists of a 3.5-nm GaAs well sandwiched between 18-nm (bottom) and 200-nm (top) $Al_{0.25}Ga_{0.75}As$ barriers on top of a 1.4-μm GaAs buffer layer. For SQW 2 the ternary Al(Ga)As barriers were exchanged by AlAs(1 nm)/GaAs(2 nm) short period superlattices (SLSs) with a 4.8-nm GaAs well in between, in order to achieve similar QW confinement energies. SQW 1 (SQW 2) was grown at a substrate temperature of 605 °C (640 °C) and an As_4-to-Ga beam equivalent pressure ratio of 16 (10). In both samples a pure (SQW 1) or modified (SQW 2) step flow growth mode was realized. The growth conditions for SQW 2 lead to an incomplete GaAs condensation resulting in a blue shift of the exciton PL from both the QW and the SLS barriers.[7] The resulting QWs can be regarded to be of state-of-the-art quality.[7] The layer sequence of SQW 3 was the same as that of SQW 2 but the growth conditions were somewhat different. In addition, a bulk Al(Ga)As and a SLS reference sample were grown under the conditions of the corresponding barriers in SQW 1 and SQW 2. Micro-PL measurements were performed with steady-state excitation at 2.18 eV using a Kr^+ ion laser with the sample temperature controlled by a He continous-flow cryostat. The PL light was analyzed by a DILOR triple spectrograph equipped with a cooled charge-coupled-device (CCD) array. With the confocal imaging of a microscope setup, an effective probe area of down to about 2 μm^2 was achieved. Time-resolved PL measurements were performed using a syncroscan streak camera system in conjunction with a Ti:sapphire laser emitting 150-fsec pulses at 1.56 eV (repetition rate of 76 MHz). For the uv excitation at 3.13 eV, a 2-mm BBO crystal was used for second-harmonic generation. The luminescence was dispersed by a single monochromator and focused onto the photocathode of the streak tube. The streak images were recorded by a cooled CCD array. The temporal resolution of the syncroscan system is 2 psec. The samples were mounted on the cold finger of a He flow cryostat.

Low-temperature (8-K) micro-PL spectra of SQW 1 are shown in Fig. 1 for an effective probe area of 2 μm^2. The PL from the GaAs QW in Fig. 1(a) is determined by excitonic recombination at 1.650 eV with a fine-structured line shape similar to those reported in Refs. 1–3. It is important to note that a similar line shape is observed for the radiative recombination in the Al(Ga)As barriers at 1.869 eV as shown in Fig. 1(b). The 5.5- and 4.5-meV broad envelopes (dashed lines) of the peaks in Figs. 1(a) and 1(b) correspond to the PL line shapes observed in conventional (macro-) PL spectra and are obtained by increasing the probe area to about 20 μm^2. As shown in Fig. 2, the envelope width and fine structure are again similar for the QW and barrier exciton PL of SQW 2 with SLS barriers; however, the PL peak envelope widths of SQW 2 (8–10 meV) are larger than those of SQW 1. The micro-PL results discussed here for SQW 1 and 2 are typical for a whole series of samples grown under various growth conditions.[7] In particular, a correlation between the envelope widths and fine structure of the barrier and QW PL peaks has been found.

Since QW thickness fluctuations have no essential influence on excitons in the barriers, the fine structure in the barrier PL is attributed to exciton localization due only to

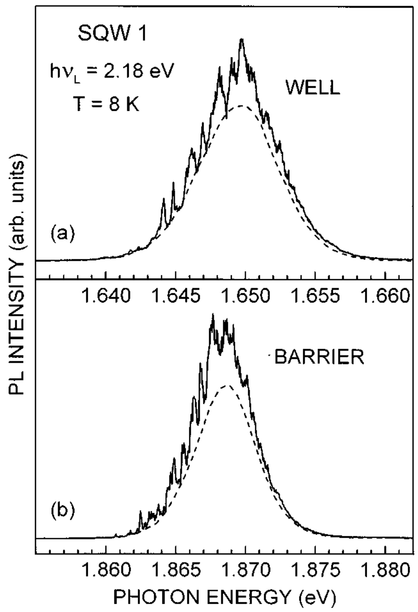

FIG. 1. Low-temperature (8-K) micro-PL (solid lines) and macro-PL (dashed lines) spectra from (a) the GaAs well and (b) the Al(Ga)As barriers of SQW 1. Excitation was at 2.18 eV. The macro-PL spectra (peak envelopes) have been reduced with respect to the micro-PL spectra by a factor of 0.8 in order to separate both curves.

Al(Ga)As composition fluctuations. Increasing the temperature up to 50 K leads to the disappearance of the fine structure in the QW PL spectra caused by a delocalization of excitons. At the same time, the barrier exciton peaks in SQW 1 and SQW 2 are quenched almost completely. This effect is

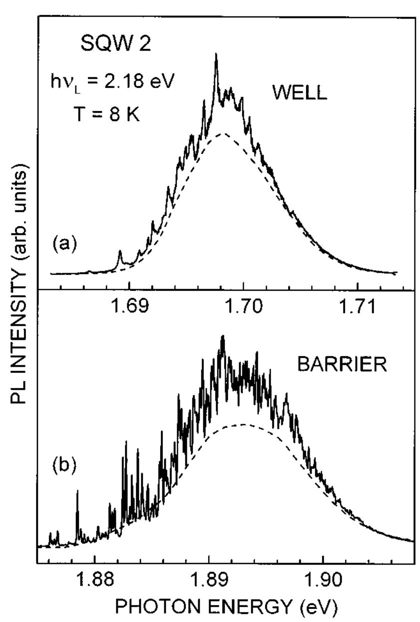

FIG. 2. Low-temperature (8-K) micro-PL (solid lines) and macro-PL (dashed lines) spectra from (a) the GaAs well and (b) the GaAs/AlAs SLS barriers of SQW 2. Excitation was at 2.18 eV. The macro-PL spectra (peak envelopes) have been reduced with respect to the micro-PL spectra by a factor of 0.8 in order to separate both curves.

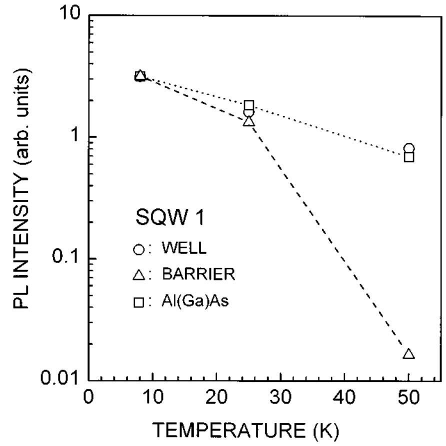

FIG. 3. Integrated intensity of PL from Al(Ga)As barriers and the GaAs well of SQW 1 together with that of a bulk Al(Ga)As reference sample as a function of temperature.

illustrated in Fig. 3 for SQW 1. Such a strong decrease of the intensity was not observed for the PL of the QW or that of bulk Al(Ga)As (see Fig. 3) and SLS reference samples. Therefore, the disappearance of the PL from the barrier cannot be explained simply by an increasing contribution of nonradiative recombination processes. Instead, this behavior must be attributed to the larger mean diffusion length of the photocreated carriers at elevated temperatures, leading to a more efficient capture in the QW.[8,9]

The question of whether or not the whole Al(Ga)As barrier region contributes to the observed PL was studied by time-resolved measurements. In order to observe time-of-flight effects we used an excitation at 3.13 eV where electron-hole pairs are photocreated only close to the surface within a penetration depth of about 10 nm. Consequently, electrons and holes are able to recombine in the GaAs QW only after diffusion through almost the whole 200-nm-thick top barrier. Therefore, if electrons and holes recombine with a constant probability during the whole diffusion path in the top barrier, the PL signal from the QW should be delayed with respect to that of the barrier. However, on the contrary, a delay of the barrier PL signal is observed. As shown in Fig. 4 for SQW 1, the maximum in the time-dependent PL intensity of the barrier [Fig. 4(b)] is delayed by about 100 psec with respect to that of the QW [Fig. 4(a)]. The effective time constant of 450 psec derived for the decay of the barrier signal has been found to be nearly the same as that for the QW signal. This is clearly seen from the constant intensity ratio shown in Fig. 4(c). The energy relaxation times of the photoexcited carriers cannot account for the different rise times of the PL intensities since they are in the picosecond range for both barrier and QW.[10] Therefore, we conclude that the PL of the top barrier is strongly suppressed in a region close to the surface.

In order to explain the delay of the PL from the barrier, we consider the mean diffusion lengths of carriers given by $l=(D\tau)^{1/2}$, where τ is the free carrier lifetime and D the diffusivity which is connected with the mobility μ by Einstein's relation $\mu=(e/kT)D$. Assuming a reasonable low-temperature ambipolar carrier diffusivity on the order of $D=1$ cm^2/sec for Al(Ga)As (Ref. 9) and a carrier lifetime in

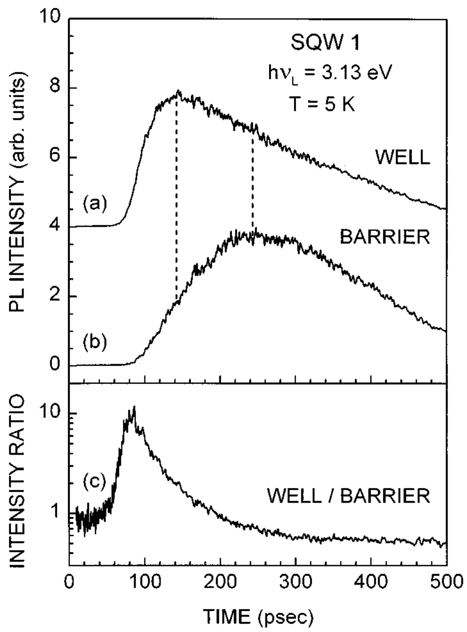

FIG. 4. Integrated PL intensity for exciton recombination (a) in the quantum well [see Fig. 1(a)] and (b) in the barriers [see Fig. 1(b)] from SQW 1 as well as (c) the ratio of both PL intensities. Excitation was at 3.13 eV and the sample temperature at 5 K.

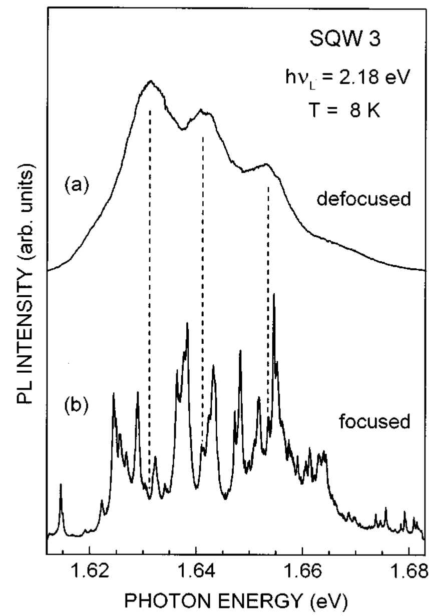

FIG. 5. Low-temperature (a) macro-PL and (b) micro-PL spectra from the GaAs quantum well of SQW 3. Excitation was at 2.18 eV.

the range of $\tau=100$ psec, we obtain a mean diffusion length of $l=100$ nm. The top barrier width $L_B=200$ nm (175 nm) of SQW 1 (SQW 2) has been designed to be just in this length scale. Therefore, we have to distinguish between the diffusion lengths of electrons (l_e) and holes (l_h). Since for electrons we expect at low temperature a much longer lifetime and a larger diffusivity than for holes, the condition $l_h \approx L_B \ll l_e$ is assumed to be fulfilled. Accordingly, the lifetime of electrons, photocreated near the surface, is much longer than the mean diffusion time into the QW, where they are captured very effectively.[9] On the other hand, a considerable part of the photocreated holes is expected to be captured by traps inside the top barrier during their lifetime at low temperature. Under these circumstances a space charge layer is built up at the QW interface due to the accumulated excess electrons, in analogy to the mechanism known as the Dember effect.[11] The induced electric field is repulsive for electrons and attractive for holes. The effective diffusion length of electrons will then be reduced by the electric field, which leads to an increasing electron density in a barrier region close to the top interface of the QW. Hence, only the localized holes in this region now find electrons as partners to form excitons and to recombine radiatively, leading the observed Al(Ga)As luminescence. Consequently, the observed time delay of the Al(Ga)As barrier luminescence corresponds to the build-up time of the electric field.

This model implies that also under the conditions of steady-state excitation at 2.41 eV the observed Al(Ga)As luminescence is restricted to a barrier region close to the top interface of the QW. This conclusion has been confirmed by preliminary numerical calculations.[12] An increase of the temperature from 10 to 50 K leads to a sufficiently enlarged hole diffusion length and hence to the observed quenching of the Al(Ga)As barrier luminescence in accordance with the above model. As mentioned above (see Fig. 3), the fact of not observing such a luminescence quenching for bulk Al(Ga)As and SLS reference samples supports our arguments. Consequently, we assign the observed fine structure in the low-temperature Al(Ga)As micro-PL to composition fluctuations in the top barrier close to the QW interface region that lead to exciton localization at minima of the barrier potential for the center-of-mass motion. Thereby, we selectively probe that region of the top barrier that is most important for confinement and localization in the GaAs QW.

The observability of the fine structure in the PL spectra of the barrier and the QW depends on the relation between the number N of exciton localization centers inside the optical probe area and our fixed spectral resolution. As shown in Figs. 1 and 2, after an increase of the probe area from 2 to 20 μm^2, which is equivalent to an increase of N by one order of magnitude, the corresponding high spectral density of narrow lines cannot be resolved anymore. The similar line shapes imply that N, i.e., the area density of exciton localization centers, is in the same range in both the QW and the barriers. Thereby one has to keep in mind that we have shown our barrier PL spectra to originate from a region close to the interface into which the QW envelope wave functions extend. It is very unlikely that the coincidence of N is just accidental. Therefore, the similar line shapes found for the QW and the barrier luminescence give evidence for the conclusion that the fine structure of the PL from the QW reflects to a large extent the distribution of exciton localization energies induced by composition fluctuations in the barrier. Concerning the confinement energy, a pure monolayer step in the QW width ($\Delta E_{\mathrm{ML}}=12$ meV) is equivalent to a homogeneous variation of the Al mole fraction x in both barriers of about $\Delta x=5\%$. The envelope peak width ($\Delta E_{\mathrm{PL}}=5.5$ meV) of the QW PL from SQW 1 [Fig. 1(a)] can be explained completely by a fluctuation of $\Delta x=\pm 2.5\%$ in one barrier that is already in the reasonable range. However, the contribution of the interface roughness still has to be taken into account. Therefore, we have to assume an even smaller degree of alloy disorder in order to explain the observed envelope peak width. In the case of samples with SLS barriers, as in SQW 2, the composition fluctuations are caused by interdiffusion at the AlAs/GaAs interfaces. The reason is a unidirectional Ga segregation in growth direction at the GaAs/AlAs interface[13] leading to intermixing of both components, Ga and Al. The degree of intermixing depends on the growth conditions.

Finally, we discuss the PL of SQW 3, which, however, does not represent the typical case of our investigated SQW structures.[7] As can be seen in Fig. 5(a), the low-temperature macro-PL spectrum consists of three peaks, which could be described by the so-called monolayer splitting. The appearance of this monolayer splitting has often been used to demonstrate the quality of QWs having large, flat growth islands. The micro-PL spectrum in Fig. 5(b), however, exhibits extremely pronounced fine structure, with the macro-PL envelope not observable anymore. Therefore, most of the excitons in the QW are strongly localized in relatively deep minima of the lateral center-of-mass potential landscape. Assuming only QW thickness fluctuations to be responsible for such a pronounced exciton localization, we would expect only one broad peak from the QW in the macro-PL spectrum according to the distribution of island sizes characteristic of the interface roughness.[5] The explanation of the transition from the micro-PL to the macro-PL spectrum is very simple if composition fluctuations in the barriers play an essential role. We just have to assume three maxima of comparable weight in the GaAs QW width distribution separated by one monolayer from each other. Averaging over a large area leads to the corresponding three peaks in the macro-PL spectrum. Their widths are then determined essentially by the distribution of exciton localization energies caused by composition fluctuations in the barriers. Thereby, we need no additional assumption concerning the lateral size distribution of growth islands at the interfaces.[6] Furthermore, the frequently reported macro-PL spectra consisting of two peaks[6] can be explained in the same way.

In conclusion, we have observed similar fine-structured line shapes of the micro-PL from both the GaAs well and the Al(Ga)As barriers of single quantum-well structures. Time-resolved measurements reveal that the observed barrier luminescence originates only from the top barrier region close to the GaAs QW interface. The results demonstrate that, besides interface roughness, composition fluctuations in Al(Ga)As barriers play an essential role for the exciton localization in GaAs/Al(Ga)As quantum wells.

Part of this work was supported by the Deutsche Forschungsgemeinschaft (DFG) by the SFB 296.

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