Quantum Dots Formed by Interface Fluctuations in AlAs/GaAs Coupled Quantum Well Structures

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We report about optical experiments on electric field tunable AlAs/GaAs coupled quantum well structures in the regime of the electric field induced Γ-X transition. Using the energetically tunable X-point state in the AlAs layer as an internal energy spectrometer and charge reservoir we are able to map out the electronic states in the neighboring GaAs quantum well in great detail. In spatially resolved and bias voltage dependent photoluminescence experiments we find sets of extremely narrow emission lines below the fundamental band gap energy of the GaAs quantum well. The new emission lines are shown to originate from natural quantum dots which are formed by well width fluctuations of the GaAs quantum well.

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The investigation of the microscopic structure of the heterointerfaces was the subject of many scientific contributions in the past. In most of those molecular beam epitaxy (MBE) grown GaAs/AlGaAs quantum wells (QW's) have been investigated; some are listed below [1–11]. Those contributions can be divided into a group of investigations on optical properties [1–9] and into a second group on atomic scale structural investigations [10,11]. Previously there was some discrepancy between the results of optical and structural investigations. Whereas by optical techniques the existence of huge monatomically smooth islands with a size of up to several μm has been claimed in high quality growth interrupted QW's [6], structural methods revealed substantial roughness and alloy fluctuations on an atomic length scale [10,11]. In more recent work, however, data from optical investigations also was found to be inconsistent with the existence of huge monatomically smooth islands [9]. Currently interface roughness with an amplitude of at least 1 monolayer (ML) in growth direction is believed to appear on a broad range of length scales reaching from atomic scale to wafer scale. In a narrow QW well width fluctuations of several monolayers will result in sizable lateral potential variations. On this basis it is justified to describe a narrow two-dimensional QW sample as a disordered array of quantum dots with arbitrary dimensions. In theoretical work well width fluctuations have been shown to strongly affect the properties of QW's in terms of eigenvalues and charge localization [12].

In the present Letter we demonstrate the existence of zero-dimensional states in narrow QW's. For our investigations we use an electric field tunable AlAs/GaAs coupled QW structure [13] to overcome the intrinsic lifetime limitations of single QW's and to benefit from resonant carrier injection into GaAs quantum dot levels. Our coupled quantum well structures are configured as heterotype n^+-i-n^+ diodes. The active pair of AlAs/GaAs layers is contained between two 400 Å wide intrinsic (i) $Al_{0.48}$-$Ga_{0.52}As$ layers. The layer sequence was grown by MBE without growth interruptions. A band diagram of the active part of the structure is shown for negative bias voltage V_B as an inset in Fig. 1. The nominal width of 30 Å has been chosen for the GaAs QW to get enough confinement energy to compensate the internal offset between the GaAs Γ conduction band (CB) edge and the AlAs X CB edge. Three different samples with AlAs layer widths of 30, 40, and 50 Å have been grown and investigated. Within the tuning range of V_B we obtain the electric field induced Γ-X transition [14]. A detailed study of the electric field induced Γ-X transition in AlAs/GaAs coupled QW's can be found in Ref. [13]. Important for this work is the huge enhancement of the exciton lifetime in the indirect regime, which can reach 500 nsec as compared to less than 500 psec in the direct

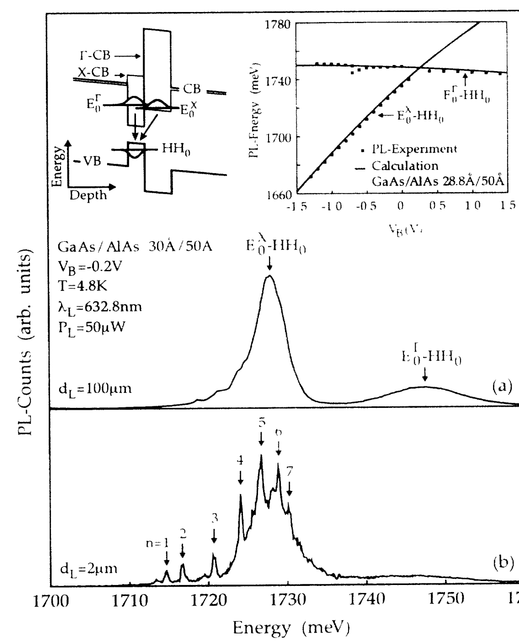

FIG. 1. (a) PL response of a GaAs/AlAs 30 Å/50 Å coupled QW structure in the indirect regime for $V_B = -0.2$ V. The diameter of the optically probed area d_L is 100 μm. A schematic band diagram of the structure and the observed PL energies as a function of V_B are shown in the inset. (b) Same as (a) only with higher spatial resolution ($d_L = 2$ μm). New narrow emission lines (labeled from $n = 1$ to 7) appear in the region of the indirect PL.

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regime [15], and also the linear Stark shift which is caused by the electric field acting on the spatially separated electron-hole system. The photoluminescence (PL) energy as a function of V_B for a GaAs/AlAs 30 Å/50 Å structure (solid squares) is shown in the second inset of Fig. 1. The solid lines represent theoretical results for excitonic transitions (no Γ-X coupling), assuming the indicated well width (GaAs/AlAs 28.8 Å/50 Å; see Ref. [13] for details). For the present work only the transitions between the individual ground states E_0^Γ-HH_0 and E_0^X-HH_0 are relevant. The Γ-X transition occurs at $V_B = 0.25$ V.

From numerical model calculations we find that a width variation of the GaAs QW by 2 monolayers (between 10 and 12 ML) will result in a 43 meV shift of the direct transition (E_0^Γ-HH_0). The major part of this shift (36 meV) appears in E_0^Γ. Since E_0^X can be tuned by more than 70 meV with respect to E_0^Γ, the X-point level E_0^X can be used as an internal energy spectrometer to map out the local energy minima of the GaAs QW in an energy range which is relevant for well width fluctuations.

Experimental evidence for the existence of zero-dimensional states in the GaAs QW is provided by different optical and magneto-optical experiments. In Figs. 1(a) and 1(b) we show PL results in the indirect regime ($V_B = -0.2$ V) for two different sampling areas. For the spectrum shown in Fig. 1(a) the laser was focused to a diameter d_L of 100 μm. The direct (E_0^Γ-HH_0) and the indirect transition (E_0^X-HH_0) is observed. The corresponding linewidth in terms of the full width at half maximum (FWHM) is 11 meV for the direct and 4.5 meV for the indirect line [16]. Whereas the direct line has a Gaussian shape as expected for the inhomogeneously broadened line of a QW grown without interruptions, a structured tail is detected on the low energy side of the indirect line. The structure in this tail appears tremendously enhanced as the laser is focused down to $d_L = 2$ μm and moved to an appropriate position in the x-y plane of the mesa diode [see Fig. 1(b)]. New extremely narrow emission lines (labeled from $n=1$ to 7) emerge from the background of the indirect line. The FWHM of those new emissions is about 0.5 meV in the observed spectrum, limited by the choice of slits in the spectrometer (the natural linewidth is about 0.2 meV). Both the narrow linewidth and the level sequence are reminiscent of an emission spectrum from a fully quantized system such as a single quantum dot [17,18]. The low energy cutoff of the narrow emission lines is about 30 meV below the position of the direct PL and 38 meV below the direct absorption edge, which corresponds to an almost 2 ML variation in well width. It can be further shown that a 1000 Å wide Gaussian-shaped potential well with a depth of about 20 meV has an almost similar level spectrum as the one shown in Fig. 1(b). The observed level spectrum changes as we choose a different position in the x-y plane of the mesa diode. In all of our samples narrow emission lines can be observed.

Results as a function of V_B from a GaAs/AlAs 30 Å/40 Å structure are shown in Fig. 2. Compared to Fig. 1(b) the observed level spectrum is more complicated. The total number of observed lines decreases with decreasing V_B. The linear Stark shift of the indirect line is still evident as a global redshift of the background from which the narrow emission lines emerge. There is, however, only negligible Stark shift on the position of the narrow emission lines. From the absence of Stark shift and the strength of the emission lines we conclude that the origin of the new lines cannot be from a real- and k-space indirect recombination process. We rather think that we observe direct transitions from zero-dimensional states in local potential minima of the GaAs QW. In general we expect to find more than one local potential minimum in an optically probed area of $d_L = 2$ μm. For small Γ-X separations carrier injection into a big variety of shallow and deep local potential minima is possible and a complex superposition of sets of narrow emission lines results ($V_B = 0.0$ V). For large separations ($V_B = -0.6$ V) only sufficiently deep potential minima can be populated and the spectra simplify. Influence on the strength of the emission lines is expected from the relaxation and injection processes. As sketched in the inset of Fig. 2, electron-hole pairs are photoexcited selectively in the GaAs QW. The electrons transfer subsequently in a fast Γ-X relaxation process [19] into the electric field tunable, long-lived E_0^X state of the AlAs QW. In this sense the X-point level E_0^X is a tunable charge reservoir which can be used to inject electrons resonantly back from the X point into local energy minima of the direct GaAs QW at the Γ point. A detailed understanding for this injection

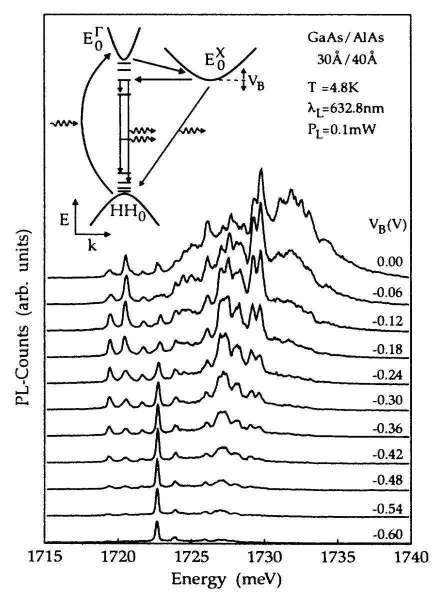

FIG. 2. PL response in the indirect regime as a function of V_B. The diameter of the optically probed area d_L is 2 μm. The relevant relaxation and recombination processes after optical excitation in the GaAs QW ($HH_0 \rightarrow E_0^\Gamma$) are shown in the inset.

process is still missing. As in the initial Γ-X relaxation the huge momentum transfer for the X-Γ process probably comes from resonant Γ-X mixing and eventually also from zone-edge phonon emission [20]. Some evidence for the participation of phonons can be seen in Fig. 2. The three energetically lowest emission lines around 1720 meV do not dominate the radiative emissions at low V_B. Instead the envelope of their intensities vs V_B seems to follow the position of the indirect line like a phonon replica, displaced by 10–15 meV.

Combining the principle of resonant charge injection with spatially resolved measurements we are able to map out the in-plane potential fluctuations. As indicated in the inset of Fig. 3 we have performed a series of spatially resolved PL measurements over an area of 45 μm×36 μm. Within this area we have recorded PL spectra with a pitch of 3 μm in the x and y directions and a laser spot diameter of about 2 μm. For each position we have plotted a PL spectrum as indicated in the inset. The displayed energy range contains only the indirect line and the new emission lines. For each given V_B the direct line appears undistorted throughout the scanned area. Part of the scanned area is masked by the gold metallization on the mesa which is shown grey in the inset. Figures 4(a) (V_B=0.1 V) and 4(b) (V_B=−0.1 V) are two representative examples from a larger series of measurements between V_B=0.2 and −0.2 V. The position and strength of the narrow emission lines in Fig. 4(a) (V_B=0.1 V) is a strong function of the in-plane coordinates. Narrow emission lines with different amplitudes can be detected over almost the entire scanned area. There exist also larger clusters with different amounts of structure in the PL spectrum. All features are strictly reproducible (even after warm-up to room temperature) and can be regarded as a fingerprint of a particular region, which is defined by a specific x-y dependence of the potential in the GaAs QW. The data are consistent with our earlier proposed picture of the GaAs QW as a disordered array of arbitrarily sized quantum dots. The spatial resolution in this experiment is twofold: First, there is the geometrical resolution given by the step size of 3 μm. In addition, there are the level spacings between the narrow emission lines which contain information about the lateral extent of the potential minima on a mesoscopic scale. Although a detailed analysis of the roughness spectrum is difficult and not the subject of this work it is still obvious that the well width fluctuations appear on a broad distribution of length scales. The amplitude of the well width fluctuations can be explored by varying the injection energy via V_B. Results for V_B=−0.1 V are shown in Fig. 3(b). As compared to V_B=0.1 V, the injection energy, which is

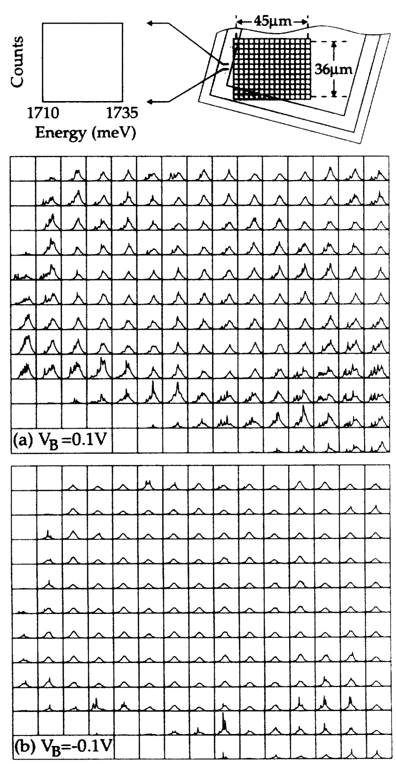

FIG. 3. Topology of the PL response for two different V_B. As indicated in the inset an area of 45 μm×36 μm has been scanned with a pitch of 3 μm in the x and y directions. A PL spectrum is plotted for each position. (a) For V_B=0.1 V huge variations of the PL response are observed, indicating substantial potential fluctuations below the injection energy. (b) At V_B=−0.1 V only a few very deep potential minima fall below the reduced injection energy.

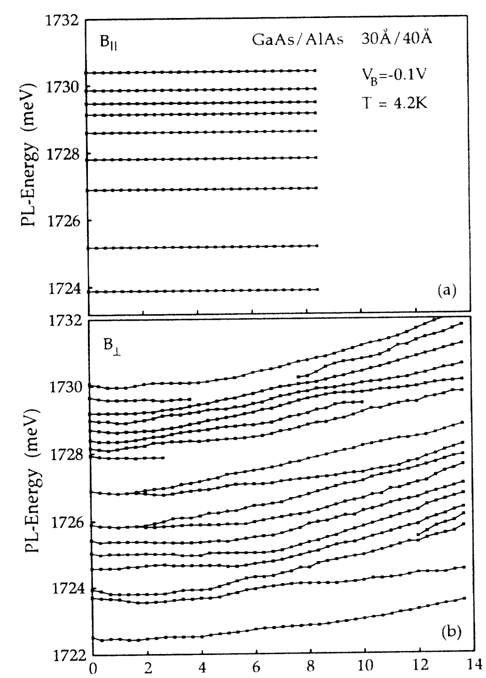

FIG. 4. Position of the narrow emission lines as a function of $B_{\parallel}$ (a) and $B_{\perp}$ (b). Different positions in the x-y plane are probed for $B_{\parallel}$ and $B_{\perp}$. Whereas for $B_{\parallel}$ the diamagnetic level shift is too small to be observed, complicated shifts, splittings, and anticrossings are observed for $B_{\perp}$.

given by the energetic position of the indirect line, is lowered by about 6 meV. At this reduced injection energy allowed energy states in the GaAs QW are harder to find. Over most of the scanned area only the undistorted indirect PL line is observed. Narrow emission lines emerge only at some selected locations. Those are the regions where the local width of the GaAs QW has maxima. In additional PL excitation measurements we found corresponding variations in the local onset of the direct gap absorption in the GaAs QW. With the detection energy on the emission line of a ground state quantum dot level we find narrow absorption lines from excited dot levels as precursors of a redshifted (up to 10 meV) two-dimensional absorption edge of the GaAs QW. This observation denotes that those electron-hole pairs which feed the narrow emission lines are photogenerated in regions with locally reduced band gap, namely, in the vicinity of the natural quantum dots.

Since our dots are by principle very asymmetric with strong confinement in the z direction (30 Å) and weak confinement in the x-y plane, their response on parallel ($B_\parallel$) or perpendicular magnetic field ($B_\perp$) has to be totally different. Experimentally we performed our magneto-optical measurements in a superconducting magnet using an optical fiber. Since the optically probed area was about 10 μm in diameter, emission lines from several local potential minima are contained in the spectra. The positions of the observed emission lines are plotted as a function of $B_\parallel$ and $B_\perp$ in Figs. 4(a) and 4(b). The probed positions on the mesa are different for $B_\parallel$ and $B_\perp$ and different sets of lines appear in both spectra. Experimentally we find negligible influence of $B_\parallel$ on the position of the narrow lines [see Fig. 4(a)]. According to $\Delta E = e^2\langle z_i^2\rangle B_\parallel^2/2m^*$, we calculate for E_0^Γ and HH_0 at $B_\parallel = 8$ T a total diamagnetic shift of only 0.2 meV, in reasonable agreement with our experimental findings (e is the electron charge, $\langle z_i^2\rangle$ the expectation value of z_i^2, i the subband index, and m^* the effective mass).

For $B_\perp$ complicated level shifts, splittings and anticrossings are observed [see Fig. 4(b)]. The complexity of the data is partly due to the fact that contributions from several potential minima are contained. If we concentrate, however, on the 3 to 4 emission lines on the low energy end of the spectrum, which originate as we think from a single potential minimum, we find qualitatively very similar behavior as theoretically predicted by Halonen, Chakraborty, and Pietilainen [21] for excitons in quantum dots in magnetic fields. For the present work we only conclude from our magneto-optical analysis that the observed quantum dots do indeed have the proposed geometry, namely, weak confinement in the x-y plane and strong confinement in the z direction.

With our experiments we have shown that the observed narrow emission lines do originate from natural quantum dots formed by well width fluctuations in the GaAs QW. The new emission lines appear up to 40 meV below the direct absorption edge, which relates to a maximum local enhancement of the well width of about 2 MLs compared to the average width. Similar or even larger variations are expected to appear in artificially made arrays of dots and wires. Potential fluctuations as reported in this work will destroy the desired overlap between the strongly peaked contributions of the density of states in those structures and will lead to unavoidable inhomogeneous broadening effects.

In summary, we have observed natural quantum dots in electric field tunable AlAs/GaAs coupled quantum well structures. The dots appear in disordered arrays and originate from width fluctuations of the GaAs QW. Using the tunable X-point state in the AlAs layer as an internal energy spectrometer and charge reservoir we have been able to populate those quantum dots by resonant charge injection from the X point. In spatially resolved and bias voltage dependent PL experiments we have observed the emissions from the quantum dots as sets of extremely narrow emission lines below the fundamental band gap energy of the GaAs QW.

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