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Optical characterization of GaAs/AlGaAs nanostructures fabricated by focussed laser beam induced thermal interdiffusion

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A novel technique is presented to modulate the effective band gap of GaAs/AlGaAs quantum well structures. Al/Ga interdiffusion on a lateral scale of about 100 nm is achieved by heating the sample locally with a focussed laser beam. Rapid thermal interdiffusion is limited to a small region underneath the laser spot center. Various lateral nanostructures have been fabricated by stepping the sample below the focussed laser beam. Wire structures with periods of about 200 nm reveal a significant peak structure and a blue shift of the photoluminescence. The strong photoluminescence efficiency allows microscopic optical characterization even of single quantum wire structures.

In order to achieve strong optical interband transitions of one- and zero-dimensional semiconductor systems, confinement of both the electrons and holes at the same spatial region is required. Commonly used etching techniques in general do not fulfill this condition. Besides some special growth techniques and strain patterning methods [1–3], also local interdiffusion within convenient GaAs/AlGaAs quantum well structures has been applied to laterally modulate the aluminium content and consequently the effective band edge energies. Bombardement by a focussed ion beam (FIB) or electron beam was used to increase local Al/Ga interdiffusion during a subsequent annealing process [4,5].

In the present contribution we describe a novel one-step technique which enables us to modulate the quantum well band gap energy directly by local thermal Al/Ga interdiffusion. This interdiffusion is induced by absorption of a focussed laser beam (FLB) which heats the sample locally to temperatures up to $T = 1000$°C. The extremely nonlinear increase of interdiffusion with local temperature is believed to cause a laterally steep, narrow interdiffusion profile in the center of the spot. Various lateral barrier profiles have been written by heating the sample point by point for adequate time durations. The interdiffusion can be controlled by micro Raman spectroscopy. The photoluminescence (PL) of the fabricated wire structures shows evidence that FLB induced interdiffusion of quantum wells indeed can generate laterally direct quantum structures of low damage and with quantization energies of the order of 10 meV.

The starting quantum well structure consists of undoped GaAs/$Al_xGa_{1-x}As$ quantum wells grown by molecular beam epitaxy (MBE). On a 3000 Å thick $Al_xGa_{1-x}As$ layer three GaAs wells of 80, 55 and 30 Å thickness and a 100 Å cap layer are separated by 200 Å $Al_xGa_{1-x}As$ barrier layers ($x = 0.35$). A 1000 Å thick Si_3N_4 film was plasma deposited onto the sample surface in order to prevent decomposition of the surface layers during laser processing. Local interdiffusion of the well and barrier layers is induced by an intensity stabilized Ar^+ laser beam ($\lambda = 5145$ Å) which is focussed by a microscope objective to a spherical spot of about 600 nm in diameter. The sample is mounted on a high precision *xyz*-translation stage which allows quasi-homogeneous writing of individual lateral structures using a

stepping mode of 25 nm step size with a position accuracy of about 10 nm. Fig. 1a schematically shows a sample which is locally interdiffused underneath the center of the focussed laser beam. Fig. 1b shows a typical radial profile of the Al/Ga interdiffusion coefficient $D(r) \sim \exp(-E_a/k_B T(r))$, ($E_a = 5$ eV), which is obtained by solving the nonlinear equation of lattice heat conduction within an $Al_xGa_{1-x}As$ sample when a FLB of total power $P = 6.6$ mW is absorbed [6]. The lateral size of the interdiffused region is strongly reduced compared to the exciting laser spot size to a total width of about 200 nm.

The microscope stage is coupled to a triple grating Raman spectrometer with a multichannel detection system. This enables us to control the laser processing parameters by in-situ micro Raman spectroscopy during patterning. The short laser penetration depth of about 100 nm and the characteristic dependence of the $Al_xGa_{1-x}As$ phonon frequencies on the Al content x allow

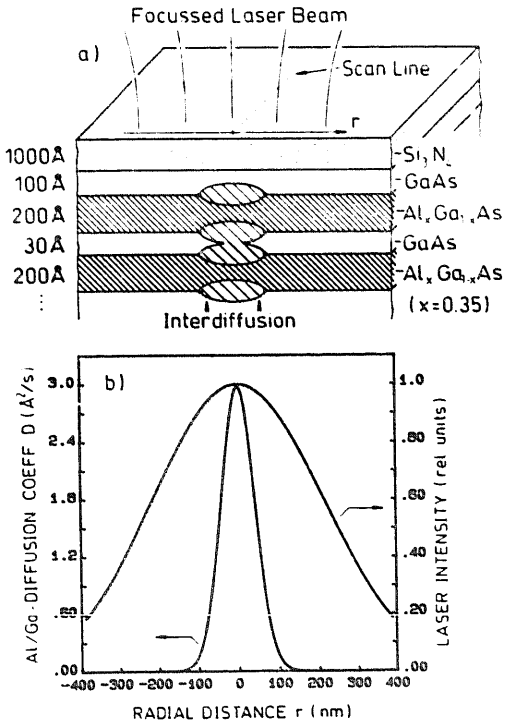

Fig. 1. (a) Schematic view of the quantum well sample with an Al/Ga interdiffused region produced by a focussed laser beam. (b) A lateral Al/Ga interdiffusion profile of about 200 nm total width was calculated from the Gaussian laser spot profile of 600 nm width (1/e).

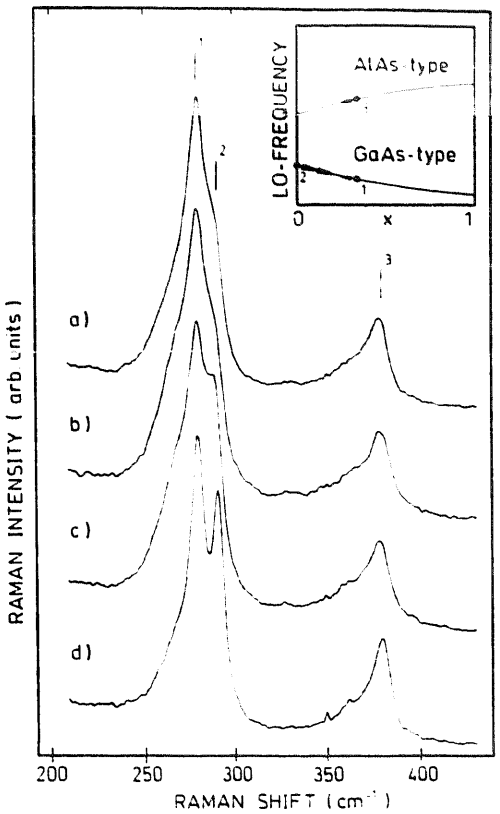

Fig. 2. In-situ Raman spectra of the GaAs/$Al_xGa_{1-x}As$ quantum well structure ($x = 0.35$) excited by a focussed Ar^+ laser beam of power P. Spectra (a), (b) and (c) were obtained with $P = 5.8$ mW and different exposure time after each scanning step of 25 nm ($\Delta t = 8.0$, 1.0, and 0.275 s, respectively). Spectrum d corresponds to $P = 2.9$ mW and 1.0 s per 25 nm step.

the observation of compositional changes of layers close to the surface. Fig. 2 shows Raman spectra which have been measured during scanning the sample underneath a laser spot of a power $P = 5.8$ mW with different delay times after each 25 nm step. Using half of the laser power (fig. 2d) no significant change of the Raman spectrum as compared to the room temperature reference spectrum is observed. With higher laser power the LO phonon of the thin GaAs layers broadens and merges with the AlGaAs phonon mode on a time scale of the order of a second. We attribute this shift of the GaAs phonon frequency to rapid diffusion of Al atoms out from the thicker barrier layers into the thin GaAs layers. An irreversible change of the local sample composition within the processed regions

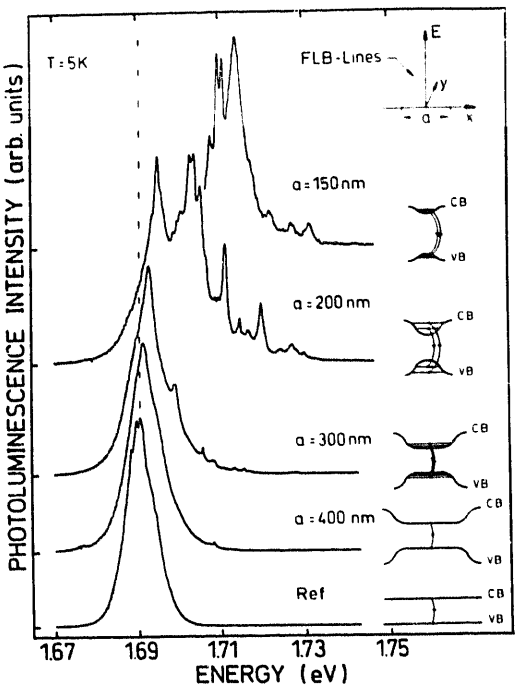

Fig. 3. Low temperature photoluminescence spectra of quantum wire structures of period a written by focussed laser beam induced interdiffusion. For PL excitation we used photon energies of $E_{Exc} = 1.76$ eV and a total power of $P_{Exc} = 10$ μW focussed to a spot of about 2 μm in diameter. A schematic view of the expected lateral conduction band and valence band modulation (CB, VB) in the various structures is given on the right-hand side.

is confirmed by subsequent Raman analysis at a strongly reduced laser intensity. An external laser power of about $P = 5.8$ mW and irradiation times of one second at each point are typical parameters used to fabricate the investigated structures.

Wire structures of different periods a have been written consecutively by FLB at constant processing parameters. The wire patterns are 8 μm $\times$ 8 μm in size and have been characterized by micro photoluminescence at low temperature ($T = 5$ K). The spatial resolution was optimized to about 1.5 μm using a cold finger He-flow cryostat with a 0.2 mm thin vacuum window and a pinhole at an image point of the PL light spot. The strongest interdiffusion effects are expected to happen within the narrowest uppermost quantum well. PL spectra of this well ($L = 30$ Å) are shown in fig. 3. For period lengths larger than 400 nm the PL is nearly unchanged compared to the unstructured regions of the sample. With decreasing period length in the wire structured region the PL maximum shifts to higher energies. There appears also some structure in the high energy tail of the PL. At a line spacing of $a = 200$ nm five predominant luminescence peaks are observed which are separated in energy by about 8 meV. Line widths as narrow as 2 meV are observed for the individual peaks. A further reduction of the period ($a = 150$ nm) leads to a shift of the luminescence to higher energy by about 20 meV. The splitting of the fine structure, however, is reduced.

The observed PL shift and splitting may be understood in terms of the effective quantum well conduction band and valence band modulation normal to the wires which is schematically shown on the right-hand side of fig. 3. While the quantum well luminescence is nearly unaffected by FLB interdiffused lines of large spacings $a \cong 400$ nm, a strong lateral modulation of the local Al content within the well is reached at periods of about 200 nm with barely interdiffused regions in between the laser scanned lines. This period compares well to the total width of the expected interdiffusion profile given in fig. 1b. For this condition the largest 1D subband separation is expected. With smaller periods the quantum well gets interdiffused even between the FLB written lines and the energy gap is rapidly increasing. Scanning the PL probe across the wire structures of narrow period $a \leq 200$ nm samples reveal some inhomogeneities. The systematic initial increase and final decrease of the peak energy separation and the increasing blue shift with decreasing period are a strong indication that lateral confinement effects are responsible for the PL peak structure. Confinement due to a sinusoidal lateral band gap modulation has been calculated for example in ref. [7]. If we scale their results to a period of $a = 200$ nm and a band gap modulation of 80 meV by the simple harmonic approximation $\hbar\omega \sim 1/a$, we get nearly equidistant electron subband separations of about 5 meV and a comparably smaller splitting of valence band states. These energies correspond roughly to the observed splitting of the luminescence line in the sample with $a = 200$ nm.

In order to avoid inhomogeneities we have also fabricated a single quantum wire structure as shown schematically in the inset of fig. 4. The wire is defined by a FLB written frame which is 340 nm × 2 μm in size. The wire structure is surrounded by a 6.2 μm × 7.8 μm sized area, where we tried to achieve uniform interdiffusion using a laser step rate of 50 nm/s. The wire defining frame as well as the outer edge of the interdiffused area are processed with a smaller step size (25 nm) in order to obtain locally a stronger and homogeneous interdiffusion. The expected band gap variation across the processed area is also shown schematically in fig. 4. The increased energy gap at the edges reduces the drift of photogenerated carriers to the minima. Spatially resolved low temperature PL spectra have been measured by scanning the sample under the probe laser spot across the structure with a step size of $\Delta x = 200$ nm. Luminescence originating from the quantum wire can be clearly separated from the surrounding as grown quantum well structure (fig. 4). The lateral width of the PL feature is determined by the experimental probe size. We want to emphasize that the PL intensity from the wire is nearly as high as from the unstructured region ($x = 0$ in fig. 4). This demonstrates that only little damage is generated within the wire by our laser processing technology. The PL of the wire reveals a slightly blue shifted asymmetrical line, whose high energy tail extends to about 20 meV above the reference PL energy. In the low energy part a splitting into discrete peaks with separations of about 2 meV is observed. Within the interdiffused area surrounding the wire a weak luminescence is detected at higher energies in some regions, like for example close to the front edge in fig. 4. We attribute these regions to local minima of the band gap, which may be caused by inhomogeneous interdiffusion. The additional blue shifted luminescence gives roughly the height of the lateral band gap modulation. The shape can be calculated from the interdiffusion profile and the actual patterning parameters. The estimated 1D subband spacings agree well with the observed peak separation in the luminescence of the single wire.

In conclusion we have demonstrated that nonlinear effects allow the direct writing of nanostructures in GaAs/AlGaAs quantum wells by optical techniques. The luminescence efficiency of laser processed structures is nearly conserved. No post processing annealing steps are required. The steep interdiffusion profile ob-

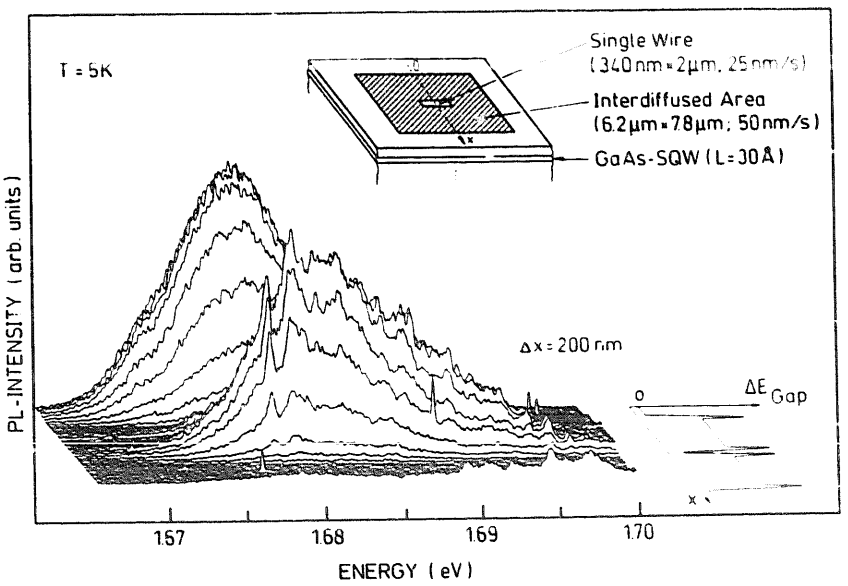

Fig. 4. Spatially resolved PL spectra at positions x ($\Delta x = 200$ nm) across a single wire structure of a size of 340 nm × 2 μm ($P_{Exc.} = 1.4$ μW; $E_{Exc.} = 1.96$ eV). The lateral shape of the structure and the intended lateral band gap modulation are shown in the insets.

tained with focussed laser beam writing results in narrow lateral confinement which opens the possibility to fabricate directly one-dimensional and, straight forward, also zero-dimensional semiconductor systems.

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