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Patterned growth on high-index GaAs (n11) substrates: Application to sidewall quantum wires

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We have recently found a new phenomenon in the selectivity of growth by molecular-beam epitaxy on patterned GaAs (311)*A* substrates to form a fast growing sidewall on one side of mesa stripes oriented along the [01−1] direction. Preferential migration of Ga atoms from the mesa top and bottom toward the sidewall forms a smooth convex curved surface profile without facets. Comparison of patterned growth on other high-index (n11)*A*&*B* surfaces shows this growth mode to be unique for GaAs (311)*A* substrates. Lateral quantum wires are realized for step heights in the quantum-size regime. Quantum confinement of excitons in the wires is demonstrated by the transition from two-dimensional to magnetic confinement with increasing magnetic field. For device applications it is important that the wires can be vertically stacked in the growth direction without increase in interface roughness and wire size fluctuations. © *1996 American Institute of Physics.* [S0021-8979(96)01619-2]

I. INTRODUCTION

In the course of lateral patterning of quantum-well structures[1–6] we have recently reported on the growth on patterned GaAs (311)*A* substrates by molecular-beam epitaxy (MBE).[7] The investigation was driven by the unique growth modes on planar (311) surfaces to naturally produce ordered quantum-wire and quantum-dot arrays in MBE and metal-organic vapor-phase epitaxy (MOVPE).[8,9] It has been found that the selectivity of growth qualitatively differs from that commonly observed on patterned GaAs (100) or (111) substrates.[10–12] On patterned GaAs (311)*A* substrates a fast growing sidewall develops on one side of mesa stripes oriented along the [01−1] direction in the sector toward the next (100) plane. A smooth convex curved surface profile is formed due to the preferential migration of Ga atoms from both sides, the mesa top and mesa bottom, toward the sidewall [i.e., opposite to the case of patterned GaAs (100) or (111) substrates]. No roughening of the growth front occurs compared to the flat parts of the mesa. Hence, very uniform thicker regions of GaAs are formed along the sidewall between thinner regions in the adjacent mesa top and bottom areas. Starting from step heights in the quantum-size regime (15–20 nm) this growth mode has been applied to realize lateral quasi-planar GaAs/(AlGa)As quantum-wire structures along the sidewall. The wires exhibit high structural perfection and uniformity, narrow emission lines in photoluminescence (PL) spectroscopy, and high PL efficiency up to room temperature.[13]

In this article we compare the growth on patterned GaAs (311)*A* substrates with that on other patterned high-index GaAs (n11) substrates. For GaAs (211)*A*, (411)*A*, (511)*A*, and (311)*B* substrates stable slow growing side facets similar to the case of patterned GaAs (100) substrates develop on both sides of all the mesa stripes oriented along [01−1] and the respective perpendicular directions. The facet formation can be related to the unique behavior of patterned GaAs (311)*A* surfaces—the only substrate orientation with a fast growing sidewall allowing the formation of lateral quantum wires. In GaAs/(AlGa)As quantum wires grown along 15-nm-high steps on patterned GaAs (311)*A* substrates, the quantum confinement of excitons is demonstrated from the diamagnetic energy shift showing a clear transition from two-dimensional confinement to magnetic confinement with increasing magnetic field. Finally, it is shown from cathodoluminescence (CL) spectroscopy that the wires can be stacked in the growth direction without any increase in interface roughness or wire size fluctuations.

II. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

The GaAs (211)*A*, (311)*A*&*B*, (411)*A*, and (511)*A* substrates were patterned by using optical lithography and wet chemical etching in H_2SO_4:H_2O_2:H_2O (1:8:40). The mesa stripes, 80 μm wide and 15–400 nm deep, were oriented along the [01−1] direction and the respective perpendicular azimuths. After cleaning the samples in concentrated H_2SO_4 the substrates were mounted side by side on the same Mo block and introduced in the MBE growth chamber. The GaAs/(AlGa)As multilayer structures (see below) were grown at 620 °C while the substrate was rotated at 6 rpm. The growth rates for GaAs and (AlGa)As were 0.5 and 1 μm/h. The group-V–to–III flux ratio was about 5. The structural and electronic properties of the layers were characterized by atomic force microscopy (AFM), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and PL and CL spectroscopies at 5 K. For PL the samples were mounted in an optical magnetocryostat. The magnetic field (from 0 to 13 T) was aligned normal to the sample surface. An Ar^+ laser was used for optical excitation with 10 W cm^{-2} power density.

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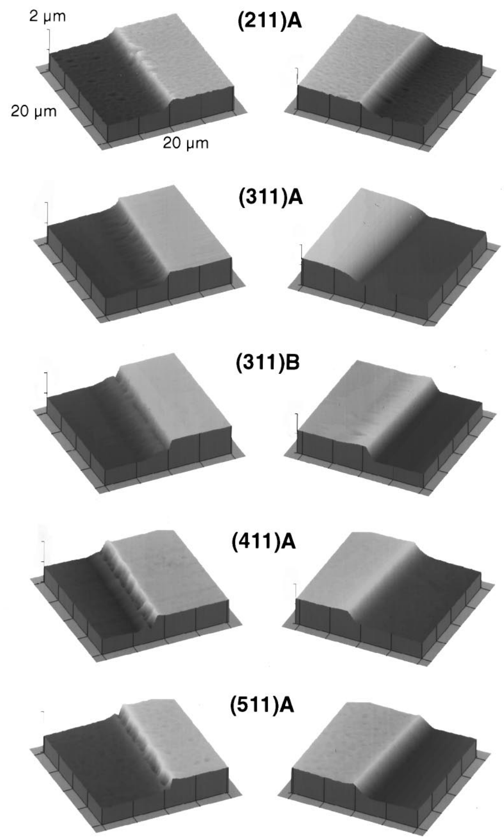

FIG. 1. Three-dimensional AFM images of the edges of the 400-nm-high mesa stripes along [01−1] after overgrowth on GaAs (211)*A*, (311)*A*, (311)*B*, (411)*A*, and (511)*A* substrates. The scanning area is 20×20 μm^2, and the full scale in height is 2 μm.

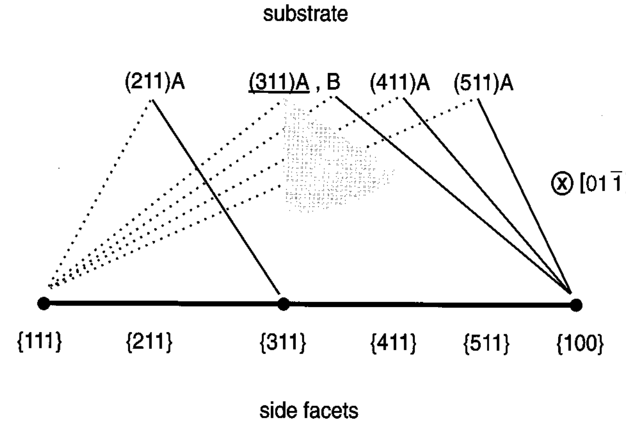

FIG. 2. Schematic diagram of the facets formed during growth on patterned GaAs (211)*A*, (311)*A*&*B*, (411)*A*, and (511)*A* substrates for mesa stripes along [01−1]. The upper line denotes the orientation of the substrate surfaces and the lower one the orientation of the facets determined from the AFM measurements in Fig. 1. The dashed lines point toward the sidewalls observed on the left-hand side from Fig. 1 and the solid lines point toward those of the right-hand side.

III. SELECTIVITY OF GROWTH ON PATTERNED HIGH-INDEX GaAs (*n*11)*A*&*B* SUBSTRATES: A COMPARISON

The growth mode on the differently oriented patterned substrates is investigated for a layer sequence comprising a 30-nm-thick GaAs buffer layer followed by five periods of 100 nm $Al_{0.5}Ga_{0.5}As$/100 nm GaAs grown over 400-nm-high mesa stripes. Figure 1 shows the three-dimensional AFM images of the surface profiles of the edges of the mesa stripes along [01−1] observed on the GaAs (211)*A*, (311)*A*, (311)*B*, (411)*A*, and (511)*A* substrates. The left-hand side shows the sidewall in the sector toward the next {111} plane and the right-hand side that in the sector toward the next {100} plane. For the GaAs (311)*A* substrate the formation of the smooth convex curved shape at the right-hand side with a sharp lower step corner is reproduced.[7] Cross-sectional SEM images show that this profile is fully developed after the growth of GaAs with a thickness comparable to the etched step height. During growth of (AlGa)As almost no change in the shape, i.e., no selectivity during growth, is observed due to the small surface migration length of Al. On the contrary, for the other substrate orientations the formation of well-defined slow growing side facets is observed along both edges of the mesa stripes along [01−1] and the respective perpendicular directions (not shown here). The evolution of the side facets in the left-hand side of Fig. 1 (in the sector toward the next {111} plane) shows pronounced roughening during growth while the surface morphology on the opposite side is rather smooth. These surface profiles are similar to those observed on patterned GaAs (100) substrates which are characterized by accumulation of material close to the upper and lower edges of the mesa forming a concave shape at the lower edge that tends to flatten the surface. This shape is due to preferential migration of Ga atoms away from the sidewall toward the mesa top and bottom to form stable slow growing side facets allowing the fabrication of V-groove or ridge quantum wires and dots.[14,15]

It is known, that lateral growth can be obtained in MOVPE or metal-organic molecular-beam epitaxy (MOMBE) in particular on patterned GaAs (111)*A*&*B* substrates due to the different chemical reactivity of the source materials on different planes.[16,17] In that case the growth on the planar surface can be almost completely suppressed. Inspecting, however, the shape of the side facets, it is again similar to that of the slow growing side facets in MBE with a concave surface profile at the bottom of the mesas. This shows that also in MOVPE or MOMBE the surface diffusion of adatoms on patterned GaAs (111) substrates is away from the side facets toward the mesa top or mesa bottom similar to the growth by MBE.[12]

A schematic illustration of the facets that form during growth on the patterned substrates at the edges of the mesa stripes along [01−1] is shown in Fig. 2. The upper line denotes the orientation of the substrates and the lower line the orientation of the side facets determined from AFM. Dashed and solid lines point toward the two opposite sides of the mesa stripe. On one side of the mesas {111} side facets develop for all samples whereas on the opposite side a {311} side facet develops for the GaAs (211)*A* substrate and a {100} facet for the (411)*A* and (511)*A* substrates. As the orientation of the facets is that of the next surface with the slowest growth rate, Fig. 2 identifies the growth rate to have a local minimum for the (311)*A* plane. The growth rate has

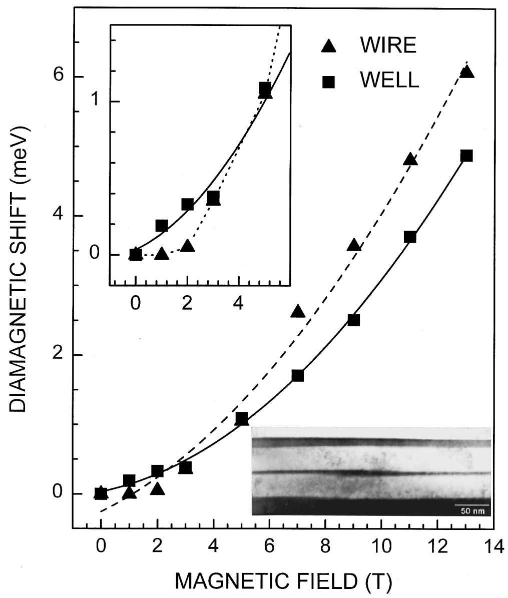

FIG. 3. Diamagnetic energy shift of the PL from the lateral quantum wire (triangles) and the quantum well in the flat part of the mesa (squares) on patterned GaAs (311)*A* substrates. The solid and dashed lines are quadratic fits to the data points. The inset shows the strong deviation from the quadratic behavior for the wire PL for $B<2$ T at enlarged scale (the dotted line in the inset is a guide to the eye). The TEM image shows the [01−1] cross section of the wire at the 15-nm-high fast growing sidewall.

to be even smaller compared to that on the next (100) plane which follows also from the appearance of {311} facets on patterned GaAs (100) substrates. In that case patterned GaAs (311)*A* substrates exhibit a fast growing sidewall on this side with a curved surface profile (there is no nearby slow growing plane to stabilize faceting) that continuously covers all surfaces between the (311)*A* close to the next {100} plane. On the other hand, on GaAs (311)*B* substrates the formation of a {100} side facet is observed. This might be due to the higher chemical reactivity of $(n11)B$ surfaces that could increase the growth rate of the (311)*B* plane compared to that of {100} planes.

IV. LATERAL CONFINEMENT IN SIDEWALL QUANTUM WIRES FORMED ALONG SHALLOW MESA STRIPES ON PATTERNED GaAs (311)*A* SUBSTRATES

Lateral quantum wires have been fabricated on patterned GaAs (311)*A* substrates along the fast growing sidewall with an etched step height of 15 nm. The layered structure consists of a 50 nm GaAs buffer layer (more than three times the step height), a 50 nm $Al_{0.5}Ga_{0.5}As$ lower barrier layer followed by a nominally 6-nm-thick GaAs quantum-well layer, a 50 nm $Al_{0.5}Ga_{0.5}As$ upper barrier layer, and a 20 nm GaAs cap layer. The quantum wire has a height of 12 nm and a lateral width of about 50 nm determined from the cross-sectional TEM image in the inset in Fig. 3. The thickness of the wire is in agreement with the red shift of the wire PL peak energy at 1.540 eV compared to that of the well at 1.602 eV in the flat parts of the mesa.[13]

To confirm the two-dimensional quantum confinement of excitons in the wire we present in Fig. 3 the diamagnetic shifts of the PL peak energies of the wire and the well in direct comparison. The PL peak of the well exhibits a quadratic energy dependence on the magnetic field in the whole range, characteristic for free excitons in quantum wells.[18] On the other hand, the PL peak energy of the wire shows a strong deviation from the quadratic behavior for small magnetic fields. Below 2 T the PL peak energy dependence of the wire on the magnetic field is very weak followed by a transition to a strong field dependence for higher magnetic fields (see inset in Fig. 3). This transition from weak- to strong-field behavior clearly demonstrates the changeover from two-dimensional quantum confinement to magnetic confinement in the quantum wire.[19] To a first approximation the transition occurs when the cyclotron diameter becomes comparable to the geometrical width of the wire. From a simple calculation the cyclotron diameter at 2 T is about 40 nm in good agreement with the wire width of 50 nm determined from TEM. The higher diamagnetic shift of the PL of the wire compared to that of the well above 2 T agrees with the increasing energy shift in thicker quantum wells expected in the regime of magnetic confinement.[18]

V. VERTICAL STACKING OF SIDEWALL QUANTUM WIRES ON PATTERNED GaAs (311)*A* SUBSTRATES

Narrow PL lines and high PL efficiency at room temperature have been observed for the present quantum wires.[13] However, for device applications it is often necessary to vertically stack the structures in order to increase the active volume. Therefore, a stack of three quantum wires (step height 15 nm) separated by 10-nm-thick $Al_{0.3}Ga_{0.7}As$ barrier layers has been grown and compared with the reference single wire structure. The buffer layer was again 50 nm GaAs and the lower and upper barriers were 50-nm-thick $Al_{0.3}Ga_{0.7}As$ layers capped with 20 nm GaAs. The nominal thickness of the GaAs quantum-well layer was reduced to 2 nm to increase the sensitivity on size and interface fluctuations of the wires. The three-dimensional AFM images of the sample surfaces containing one or three wires show the same smooth convex curved surface morphology (see the inset in Fig. 4 for the multiple quantum-wire structure).

The CL spectra in Fig. 4 excited on the sidewall of the single [Fig. 4(a)] and multiple [Fig. 4(b)] quantum-wire structures each show two lines from the wire and the adjacent well. The red shift of the CL peak energy of the wire compared to that of the well indicates a wire thickness of about 4 nm when lateral confinement effects are neglected. Hence, the selectivity of the growth mode across the edge is confirmed for the 2-nm-thick GaAs layer. The difference in the peak energy positions of the single and multiple wire structure is attributed to the higher step height of the single wire structure of 30 nm changing the shape of the curved surface profile as the total step height has to be maintained during growth. In the flat parts of the mesa the CL line positions of the single and multiple quantum wells are almost the same. The linewidth of the CL from the well corresponds to interface fluctuations of one monolayer. Most surprisingly, however, the CL linewidth of the multiple quantum wire is not enhanced compared to that of the single quantum wire.

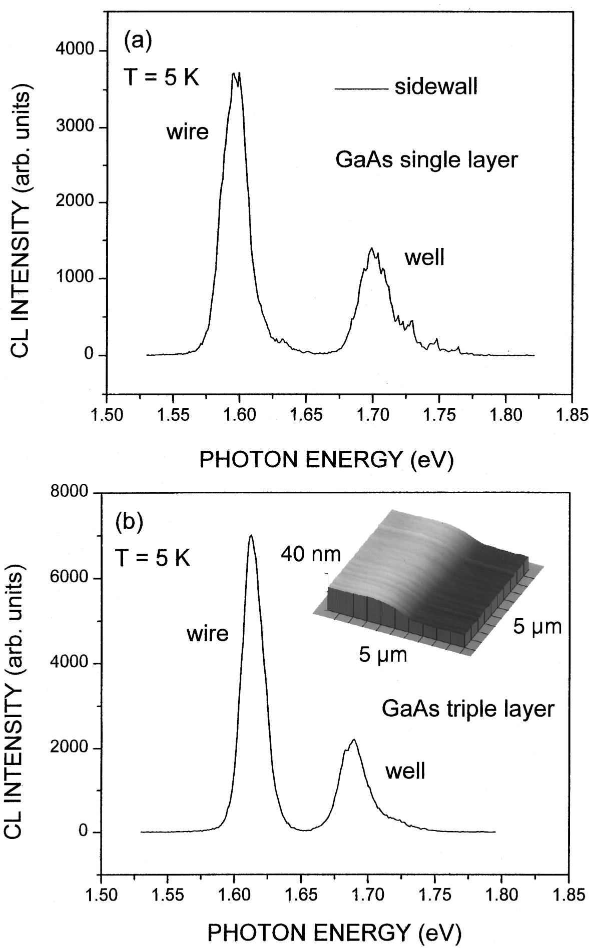

FIG. 4. CL spectra at 5 K excited at the fast growing sidewall of (a) the single and (b) the stacked multiple lateral quantum-wire structure on patterned GaAs (311)*A* substrates. The nominal GaAs layer thickness is 2 nm. The inset shows the three-dimensional AFM image of the 15-nm-high sidewall of the multiple quantum-wire structure.

(for comparison, the CL from the stacked wells on the opposite slow growing sidewall of the mesa stripe shows three distinct peaks at positions similar to the peak on the mesa top or bottom). Hence, for a given step height, a considerable number of quantum wires can be stacked in growth direction without any increase of interface and size fluctuations. This behavior suggests a self-limiting growth mechanism with well-defined lateral growth along the corner at the bottom of the surface profile.

VI. CONCLUSION

In conclusion we have investigated the growth on patterned GaAs (211)*A*, (311)*A*&*B*, (411)*A*, and (511)*A* substrates by molecular-beam epitaxy. For all substrates besides GaAs (311)*A*, stable slow growing side facets evolve along the edges of mesa stripes oriented along the [01−1] direction and the respective perpendicular azimuths. Only on patterned GaAs (311)*A* substrates a fast growing sidewall exists on one side of mesa stripes along [01−1] forming a smooth convex curved surface profile during growth. This growth mode has been applied to the formation of very uniform lateral quantum-wire structures along the sidewall. The two-dimensional quantum confinement of excitons in the wires has been demonstrated from the diamagnetic shift of the wire photoluminescence peak energy compared to that of the well on the mesa top or bottom. The quantum wires have been vertically stacked in the growth direction without an increase in interface roughness and wire size fluctuations.

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