Adatom migration effects during molecular beam epitaxial growth of InGaAs/GaAs quantum wells on patterned substrates with vertical sidewalls: Blue shift in luminescence spectra

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(Received 20 November 1995; accepted for publication 19 December 1995)

We have studied the blue shift in photoluminescence emission energy of pseudomorphic InGaAs/ GaAs quantum wells grown on patterned (001) GaAs substrates with grooves and trenches having vertical sidewalls made by dry etching. Dependence of the blue shift, which can be as large as 51 meV, on the direction, feature size, and the etch depth of the patterns as well as the thickness of the buffer layer was observed, and is explained by the altered migration behavior of the adatoms. © 1996 American Institute of Physics. [S0003-6951(96)04308-X]

Molecular beam epitaxial (MBE) growth on nonplanar patterned substrates has been studied with particular attention to its application in defect reduction in large lattice mismatched systems, realization of low dimensional (quantum wire and quantum dot) structures, and lateral band-gap variation in a single growth step.^{1–5} Due to the large difference in migration lengths of Ga and In on GaAs, growth of the ternary alloy InGaAs exhibits lateral compositional variation when grown on patterned GaAs substrates.^{5,6} There are reports of MBE growth on GaAs substrates patterned with (n11) sidewalls obtained by wet etching.⁴⁻⁶ The angled sidewalls in this case act as sources of excess In due to the nonisotropic preferential migration of adatoms away from the higher index (n11) planes. This gives an increased In composition on the patterned regions when the dimensions of the pattern are of the order of the In migration length.⁵ This technique, however has the disadvantage that it is difficult to control the change in In composition accurately due to the evolution of multiple facets during MBE growth.³

We have recently shown⁷ that growth on substrates patterned with vertical sidewalls, on the other hand, produces a decrease in In composition. The blue shift in the band-gap thus obtained is found to be accurately reproducible due to the availability of high quality dry etching techniques such as reactive ion etching (RIE) with or without an electron cyclotron resonance (ECR) source. In this letter, we report the dependence of the observed blue shift on the dimensions of the ridges and grooves as well as the etch depth. For device structures (such as lasers) it is necessary to grow thick layers (cladding layers) below the active region on the patterned substrates. This produces some smoothening of the patterns before the growth of active layers is initiated, which is found to reduce the energy shift obtained by this technique. However, we also show that the effect of smoothening on the In migration can be compensated, to a certain extent, by increasing the etch depth.

GaAs (100) substrates were patterned with RIE in a dual chamber PlasmaTherm system with BCl₃:Ar (11 sccm:21 sccm) at 15 mT and 25 W of rf power. Ridges and trenches of dimensions varying from 5 to 25 μ m were etched to

depths of $1-3 \mu m$. A chemical etch with H₂SO₄:H₂O:H₂O (1:8:80) was done to remove the process induced damages, followed by a 30 s dip in HCl:H 2O (1:1), before the samples were introduced into a Varian GEN II MBE system. Two different structures were grown on these patterned substrates. The first is a single 60 Å In_{0.2}Ga_{0.8}As quantum well (QW) with 0.2 μ m GaAs barrier layers on both sides, grown without a buffer layer. The second type of samples were grown to investigate the effect of smoothening of the patterns by a thick pregrowth. In these, buffer layers of 1 μ m Al_{0.3}Ga_{0.7}As were grown before growing the In_{0.2}Ga_{0.8}As/GaAs QW structure as described above. Particular care was taken to maintain the same growth rate and V/III ratio for all the samples to eliminate the contribution by the flux ratio dependent migration effect.⁸ The lateral band-gap variation caused by the compositional variation was studied by measuring the photoluminescence (PL) spectra. These spectra were measured at 18 K with a 488 nm Ar⁺ laser excitation source, a 0.75 m scanning spectrometer, liquid nitrogen cooled photomultiplier, and lock-in amplification. The energy shifts for the patterned regions were measured with reference to the excitonic emission from an unpatterned region of the same sample. The excitation laser beam was focused to a spot size of the order of the size of the patterns (~10 μ m). The incident power density was ~300 W/cm².

Photoluminescence spectra obtained at 18 K from the patterned region (Fig. 1) showed a blue shift in the excitonic peak compared to that from the unpatterned region. One important observation here was that the PL emission peak



FIG. 1. Schematic of a typical patterned substrate on which different structures explained were grown.

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FIG. 2. Dependence of energy blue shift on the lateral dimensions of the patterns for single 60 Å $In_{0.2}Ga_{0.8}As$ QW grown on patterned GaAs substrates (a) without a buffer layer and (b) with 1 μ m $Al_{0.3}Ga_{0.7}As$ buffer layer.

shifted to higher energy without any noticeable change in either the intensity or the linewidth. However, there was a degradation in the linewidth of the PL emission from the grooves, possibly due to the RIE induced damage to the surface being etched. This strongly suggests that good device quality material with lateral band-gap variation can be grown on top of the ridges rather than in the grooves. The energy shifts of the PL emission peak thus obtained depend on the dimensions of the patterns. Figure 2(a) depicts the variation of the energy shift with lateral dimension of the ridges and grooves aligned in two perpendicular directions. It is observed that the blue shift increases with decreasing width and spacing for both ridges and grooves. We have observed a maximum shift of up to 51 meV for ridges with the minimum width used in these experiments (10 μ m). In fact, we have earlier reported a shift of 95 meV (Ref. 7) for square grooves (8 μ m×8 μ m), in which case the patterning effects are present in both directions. It is interesting to note that larger energy shifts are obtained for ridges along [011] direction compared to ridges along [011] direction. Additionally, we have observed faceted growth when the ridges are oriented along the [011] direction. Due to this strong faceted growth there is a tendency for In migration towards the center of the ridge from the higher index facets, just as in the case of angled (n11) sidewalls.⁴ This opposes a net In migration towards the sidewalls, thereby reducing the observed blue shift.



FIG. 3. Scanning electron micrograph of the stained cross section of the structure grown on patterned substrate having sidewalls with $\theta > 90^{\circ}$.

The cause for the blue shift of PL emission peak obtained from the ridges and trenches with vertical sidewalls is contrary to that for the red shifts reported in the literature for patterns with angled sidewalls, 4-6,8 in terms of adatom migration. In the latter case, there is preferential migration of adatoms away from the higher index planes. Thus, the angled sidewalls act as effective sources for net In migration towards the center of the ridges and trenches. In the case of vertical sidewalls, ideally there is no component of flux available for growth on sidewalls ($\cos \theta$ effect). However, we have noticed that during MBE there is growth on the vertical sidewalls as well. In order to confirm that this growth is indeed due to the effective migration of adatoms towards the sidewalls, we have carried out growth on patterned substrates with sidewalls angled inwards ($\theta > 90^\circ$), obtained by a combination of dry and wet etching. A scanning electron micrograph (SEM) of the stained cross section of this grown structure is shown in Fig. 3. Clearly there is growth on the sidewall, although it was shadowed from the beam fluxes. There is also an enhanced growth near the edges of the ridge because these edges provide kink sites for the migrating adatoms. The effect of this net mass transport is observed to be limited to a lateral dimensions of $\sim 1 \ \mu m$ for Ga because the migration length of Ga over GaAs is about 1 μ m.⁹ However, since the migration length of In over GaAs is about 25 μ m (Ref. 6) under typical MBE growth conditions, we can expect a net In migration towards sidewalls when the dimensions of the patterns are of the order of 25 μ m. This gives a decrease in In composition and a corresponding increase in band gap for the material grown on the ridges and grooves. Thus the vertical sidewalls act as effective sinks for the migrating adatoms due to the dual effect of growth on sidewalls and enhanced growth at the edges of the patterns.

In order to utilize the observed blue shift in the monolithic integration of devices such as lasers and modulators, it is necessary to consider the effect of the growth of thick layers, such as the cladding layer of a laser, below the active layers. Figure 2(b) shows the lateral energy shifts in these samples for two different etch depths. Compared to Fig. 2(a), the blue shifts are indeed reduced by the thick growth. This



FIG. 4. PL spectra from the two sections of a ridge with stepped ridge widths.

is due to the smoothing of the edges and the overall filling of the patterns during growth of the 1 μ m Al_{0.3}Ga_{0.7}As layer. However, it is observed from Fig. 2(b) that the smoothening effect can be partially overcome by having a deeper etch for the patterns, in which case the pattern filling effects are reduced. We can thus infer that structures with a wide range of total thickness for lasers, modulators, detectors and waveguides, with material energy band gaps close to each other, can be grown simultaneously on the substrates patterned with vertical sidewalls. Figure 4 depicts the photoluminescence spectra obtained from such a two-section ridge with stepped ridge widths. The measured linewidths of the emission peaks are 3.5 meV. These results predict that one of the most stringent requirements, that of narrow linewidths for both the sections, can be satisfied by this approach. In conclusion, we have shown that the observed blue shifts obtained in the PL emission peak from $In_{0.2}Ga_{0.8}As/GaAs$ QW on the ridges and grooves of patterned (001) GaAs with vertical sidewalls depend not only on the lateral dimensions of the patterns but also on the thickness of the buffer layer and etch depth of the patterns. The energy shifts are obtained in this technique without any degradation in the intensity or the linewidths of the PL excitonic peaks. Growth on the sidewalls and enhanced growth near the edges, which act as sinks for the migrating adatoms, are believed to be responsible for the decrease in the composition of InGaAs on the ridges and in the grooves.

This work is supported by the Advance Research Projects Agency (COST) MDA 972-94-1-0004 by the Office of Naval Research under Grant N00014-90-J-1831 (027494) and the Army Research Office under Grant DAAL 03-92-G0109.

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