

Surface smoothing of GaAs microstructure by atomic layer epitaxy

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We report a method to smooth the rough surface of GaAs microstructures. This method is based on the nucleation process for atomic layer epitaxy which involves the self-limiting two-dimensional (one-monolayer) island formation. The method has been applied successfully to smooth the (111)A surfaces of chemically etched V-grooved GaAs structures as well as the (110) and (111)B side walls of selectively grown GaAs stripe structures. © 1999 American Institute of Physics. [S0003-6951(99)01607-1]

Microstructure formation of semiconductor devices has been primarily developed with the help of photolithography and etching.¹⁻⁴ Surface and edge roughnesses, being unavoidable in the nanostructure/microstructure processing, are essential problems that degrade physical properties and device performance of the structures. One of the typical examples is that the edge roughness of mask layers formed by the etching process results in the striae on side facets.⁵ The striae usually lead to an increase in the threshold current of Fabry–Perot mirror lasers because of the reduction of optical reflectivity.

In this letter, we are concerned with smoothing the rough surface of GaAs microstructures by atomic layer epitaxy (ALE). Advantages of ALE are: (i) self-limiting two-dimensional island formation (Fig. 1); (ii) excellent selectivity among different surfaces;^{6,7} (iii) good thickness controllability; and (iv) good uniformity.⁸ We have inferred that these features may be applicable to recover the damaged surfaces of microstructures without significantly changing the shape and size of original structures.

The concept of the surface smoothing method is schematically illustrated in Fig. 2. To be simple, let us assume that the rough surface consists of x and y planes. Let us choose the specific ALE growth condition so that the growth rate on the x surface is extremely lower than that on the y surface. We expect that the growth on the x surface proceeds very slowly, whereas the growth on the y surface occurs relatively fast until the flat x surface is established automatically. Additional supply of source gases will not cause any further growth on the x surface. In this method, the best result would be obtained when the growth selectivity is perfect between x and y surfaces. Moreover, the crystal nucleation has to proceed with the formation of self-limiting two-dimensional (2D) islands. Observation by atomic force microscopy (AFM) have verified that ALE of GaAs is primary driven by the formation of 2D islands, as shown in Fig. 1.

We first describe the experimental results for smoothing the (111)A surfaces in V-grooved GaAs structures. They were fabricated by selective wet etching through the stripe

windows on a SiO₂ mask layer.⁹ A schematic illustration of the V-grooved structure is shown in Fig. 3(a). The width and depth of typical V grooves were 10 and 7 μm , respectively. Figure 3(b) shows the bottom part of the as-etched structures in which we can clearly observe a number of etch pits and ridge striae on the (111)A side walls. The striae are attributed to the edge roughness of a patterned photoresist polymer layer, which is inevitable for the conventional photoresist materials. Figure 3(c) shows the bottom part of the V groove after growing a thin GaAs layer by the conventional metal-organic vapor phase epitaxy (MOVPE) process. Here, the growth conditions are as follows: the substrate temperature $T_s=600^\circ\text{C}$, the reactor pressure $P_r=100$ Torr, and trimethylgallium (TMGa) and arsine (AsH₃) supply rates of 3.7×10^{-2} and $3.0\times 10^1 \mu\text{mol/s}$, respectively. The growth time was 8 min, which corresponds to the layer thickness of 200 nm on (001)GaAs surface. As seen in Fig. 3(c), the surface roughness appears to be emphasized. This could be a natural consequence of the step-flow mode of MOVPE process. Moreover, the sharp bottom profile of the V groove is rounded, which often occurs for samples obtained by MOVPE.⁴ On the basis of the experimental results, we conclude that MOVPE growth would not meet the demand for smoothing the rough surface.

In contrast to the result obtained by MOVPE, the surface smoothness in V-grooved structures is clearly improved by

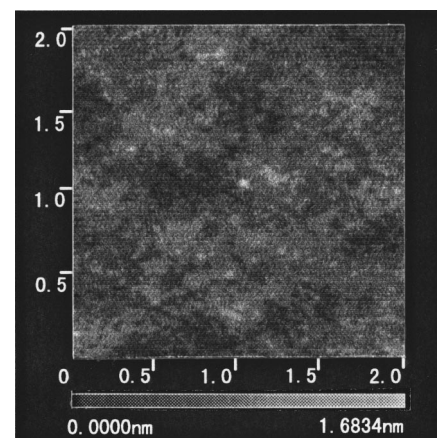


FIG. 1. Atomic force microscopy plan-view image of the GaAs(001) surface grown by ALE at $T_s=480^\circ\text{C}$, showing 2D-island formation.

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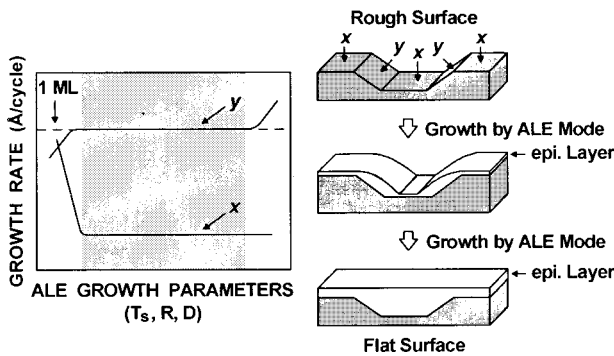


FIG. 2. Schematic illustration showing the method to smooth the rough surface by using ALE growth process.

applying the ALE process, as shown in Fig. 3(d). The ALE conditions used for this experiment are $T_s = 480\text{ }^\circ\text{C}$, $P_r = 10\text{ Torr}$, and TMGa and AsH₃ supplies of 1.5×10^{-1} and $3.0 \times 10^1\text{ }\mu\text{mol/s}$ for 3 and 10 s, respectively, with 3 s of hydrogen purge between each source supply. The ALE process is repeated 707 times, which corresponds to the layer thickness of 200 nm on GaAs(001) surface. We discuss the reason for choosing this particular T_s in the later paragraph referring to Fig. 4. As seen clearly in Fig. 3(d), unwanted surface roughness shown in Fig. 3(b) vanishes almost completely after the ALE process. Besides this improvement, a

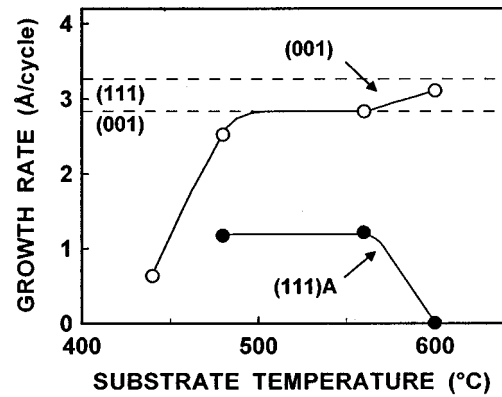


FIG. 4. Growth rates per ALE supply cycle on (001) and (111)A GaAs surfaces as a function of substrate temperature.

(001) plane is spontaneously developed at the bottom of the V groove with sharp edges at the intersects between (001) and (111)A surfaces. The observed results indicate that smoothing the surface of GaAs microstructure is achieved by the ALE process.

Figure 4 shows the dependence of GaAs growth rate on substrate temperature (T_s) for (001) and (111)A surfaces. The dashed lines in the figure represent the growth rate of one monolayer (ML) per ALE cycle for (001) and (111)

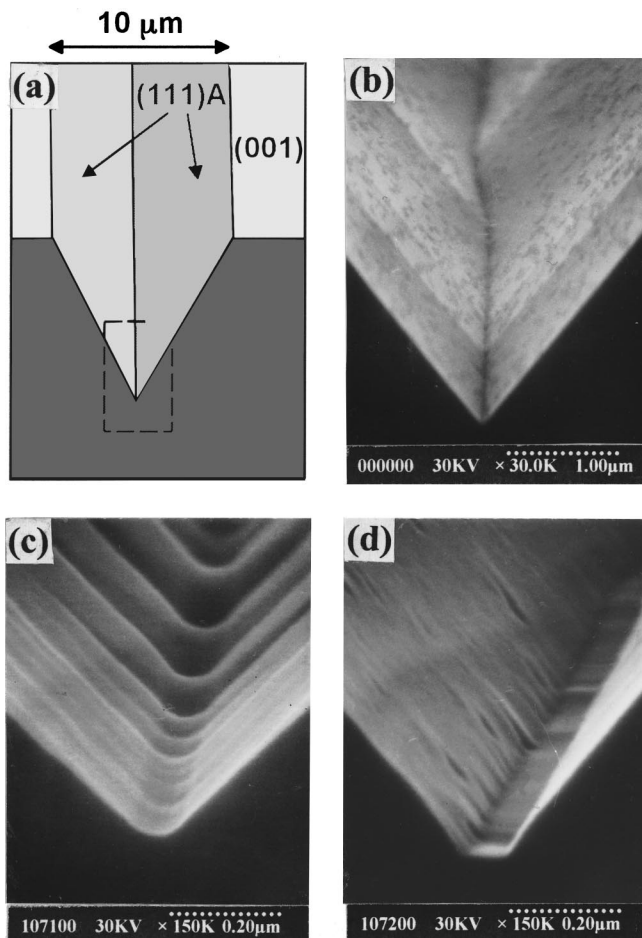


FIG. 3. (a) Schematic illustration of a V-grooved GaAs surface, (b) SEM images of a V-grooved GaAs surface after wet chemical etching, (c) and (d) SEM images of V-grooved GaAs surfaces after growth by MOVPE and by ALE, respectively.

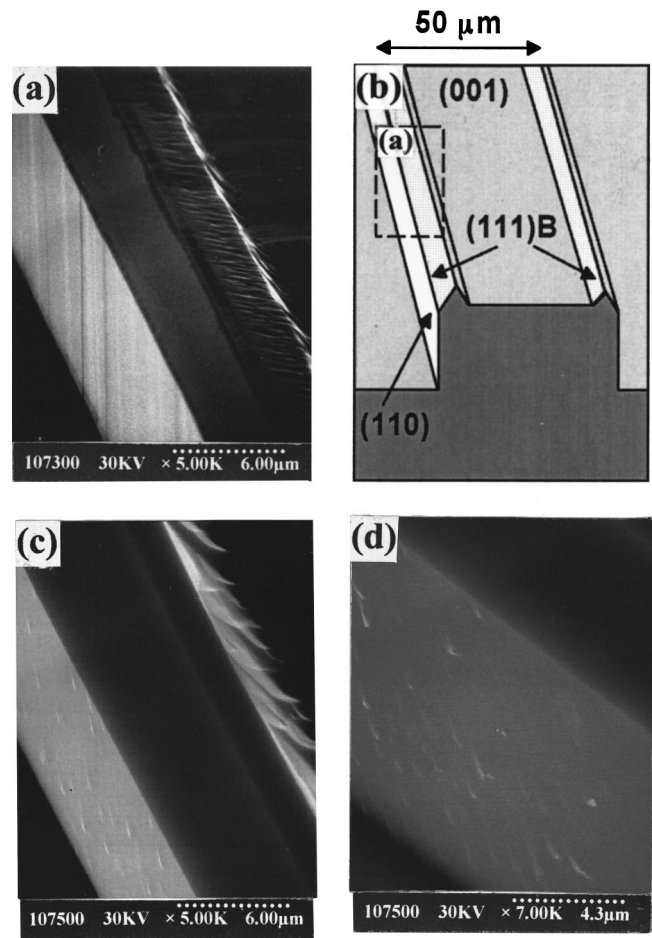


FIG. 5. (a) SEM image of a side wall of a GaAs stripe structure grown by selective area MOVPE, (b) schematic illustration of the structure, (c) and (d) SEM images of the structures after ALE smoothing method with magnification of $\times 5000$ and $\times 7000$, respectively.

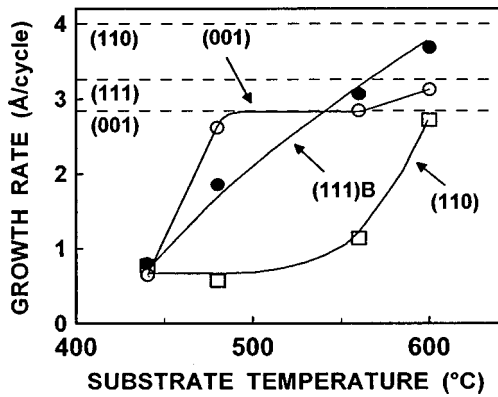


FIG. 6. Growth rates per ALE supply cycle on (001), (111)B, and (110)GaAs surfaces as a function of substrate temperature.

planes. For the ALE growth on the (001) plane, the growth rate increases with T_s up to $T_s \approx 500^\circ\text{C}$, saturates at 1 ML/cycle in $T_s = 500\text{--}570^\circ\text{C}$, and starts increasing again at $T_s > 570^\circ\text{C}$. On the other hand, the growth rate on the (111)A plane is about one third of that of the (001) plane, and stays almost constant at $T_s = 480\text{--}570^\circ\text{C}$. It appears to decrease with further increasing the T_s . On the basis of these data, we have chosen the substrate temperature of $T_s = 480^\circ\text{C}$ for the smoothing experiment with the expectation that the growth would stop automatically when a well-defined flat (111)A surface is established on the V-grooved side wall. As demonstrated in the previous paragraphs and Fig. 3, this turns out to be the case in this particular experiment.

We now show the second example of surface smoothing in Figs. 5(a)–5(d). This experiment was carried out for side walls of the stripe structure grown selectively by conventional MOVPE at $T_s = 700^\circ\text{C}$ with TMGa and AsH₃ supply rates of 3.7×10^{-2} and $1.5 \mu\text{mol/s}$, respectively. In addition with the (110) side walls, triangular ridge structures with (111)B surfaces were spontaneously developed at the edge parts of the GaAs stripes [Fig. 5(a)]. The structure, as a whole, is schematically illustrated in Fig. 5(b). Many wavy striae exist on (110) surface [Fig. 5(a)], and the (111)B surface is also not completely flat, showing irregular holes particularly at the top of the ridge. All these features are originated from the edge roughness existing in window regions on a SiO₂ mask layer.

In order to recover the defective surface, we have applied the ALE process to smooth both (110) and (111)B surfaces. Figure 6 shows the GaAs growth rate on three different surfaces as a function of T_s , obtained under TMGa and AsH₃ supply rates of 3.7×10^{-2} and $3.0 \times 10^1 \mu\text{mol/s}$, with supply times of 2 and 10 s, respectively. The point here is to find the T_s at which the growth rate on the (110) surface is relatively slower than those on the other surfaces. T_s

$= 480^\circ\text{C}$ satisfies this condition in that 1-ML self-limiting ALE growth mode is almost realized on (001) surface while the growth rate on (110) is less than 1 Å per cycle. The growth rate on (111)B plane is in between those on the (001) and (110) surfaces.

Figures 5(c) and 5(d) are birds-eye scanning electron micrograph (SEM) images of the stripe structure after applying the ALE smoothing process at $T_s = 480^\circ\text{C}$. The ALE process is repeated 100 times. As seen clearly in Figs. 5(c) and 5(d), both wavy striae on the (110) surface and pinholes on the (111)B surface disappear after the smoothing process. The wavy surface features, being formed originally by MOVPE process, are not repaired by this process. Also, the inner (111)B plane still remains to be somewhat rough. We infer that the roughness of these defective structures were too large to smooth out completely by the single smoothing process. Applying another ALE process with different conditions would result in further improving the surface roughness.

In conclusion, we have demonstrated a method to smooth the surface of GaAs microstructures by using ALE. We believe that the method is based on the self-limiting 2D-island nucleation, being unique for the ALE growth, and may be applicable to the surfaces of other III–V compound semiconductors such as phosphides and nitrides.

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⁹A 200-nm thick SiO₂ mask layer was first deposited by chemical vapor deposition on a (001)GaAs substrate. It is then patterned by conventional photolithography and wet chemical etching (H₂SO₄:H₂O₂:H₂O = 1:1:100) to yield the V-grooved structures along the [110] direction. Finally, the oxide layer was removed by BHF solution. The surface orientation of V grooves is (111)A.