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Annealing of InGaAlAs digital alloy studied with scanning-tunneling microscopy and filled-states topography

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We have investigated the structural properties of as-grown and annealed (750 and 800 °C) digital alloy InGaAlAs (λ = 1.3 μm) laser structures by cross-sectional scanning-tunneling microscopy. We show that it is possible to resolve the digital alloy period in the as-grown sample and the 750 °C annealed sample. The 800 °C annealed sample did not show the digital alloy period because of intermixing of the digital alloy. The 750 °C annealed sample showed only slight intermixing. The barrier/well interface roughness for the as-grown and the 750 °C annealed samples was the same. Annealing at 800 °C showed large barrier/well interface roughness and lateral composition modulation due to the phase separation of InGaAs/InAlAs alloys. © 2003 American Institute of Physics. [DOI: 10.1063/1.1555265]

Digital InGaAlAs alloys, lattice-matched to an InP substrate, have been used successfully to construct laser structures without the need for additional source cells or laborious changes of cell temperature during growth with molecular-beam epitaxy. With this technique, InGaAlAs alloy layers are formed by sequentially depositing (fractions of) monolayers of separate alloys, which can be either ternary (InGaAs/InAlAs) or binary (InAs/GaAs/AlAs). The resulting deposited layers have on average, a quaternary composition.

A possible drawback of digital alloys, however, is the introduction of many heterojunction interfaces of the short-period superlattice (SPS). Probably more important is the fact that the optimum growth temperature $T_{GR}$ in view of the congruent sublimation temperature ($T_{CS}$) of the ternary InGaAs alloy, is lower than the $T_{CS}$ of the InAlAs alloy. To prevent degradation of the InGaAs alloy, the growth temperature is limited to about 510 °C. This leads, however, to the incorporation of nonradiative recombination centers in the unintentionally low-temperature-grown InAlAs alloy. It has been suggested that rapid thermal annealing (RTA) removes most of the nonradiative recombination centers.

We report cross-sectional scanning-tunneling microscopy (X-STM) measurements of as-grown and annealed digital alloy InGaAlAs 1.3-μm laser structures. Room-temperature photoluminescence (PL) measurements have previously shown that after annealing at a temperature of $T_{RTA} = 620$ to 750 °C, there is a huge increase of the PL intensity and a small (10-meV) blueshift of PL peak energy. At $T_{RTA} = 800 °C$, PL measurements have shown degraded PL intensity. The blueshift has been attributed to slight intermixing of gallium and aluminum in the InGaAs/InAlAs SPS interfaces. The degraded PL intensity of the $T_{RTA} = 800 °C$ annealed structure has been attributed to roughening of the barrier/well interfaces. With X-STM, we have found direct evidence for the smoothing of the SPS at and above $T_{RTA} = 750 °C$ and strong roughening of the multiple quantum well (MQW) interfaces at $T_{RTA} = 800 °C$. We did not find any evidence for nonradiative recombination centers (vacancies) with X-STM because of the low density of these features and the difficulty of distinguishing them in the alloyed material.

The digital alloyed InGaAlAs layers were grown on an epiready n-InP substrate (see Fig. 1). The layers were grown at a growth temperature of 510 °C. Details of the grown structure and growth conditions are well described elsewhere. The wafer was covered with a 1500-Å SiO$_2$ layer to prevent re-evaporation of arsenic during RTA. Annealing was done in a nitrogen atmosphere for 30 s at 750 and 800 °C.

STM measurements have been performed in an UHV chamber with base pressure $<5 \times 10^{-11}$ Torr on the UHV-cleaved (110) cross-sectional surface. Tips are prepared by electrochemical etching of polycrystalline tungsten wires and are treated in the vacuum with a self-sputtering technique.

![FIG. 1. Schematic of the sample structure.](https://example.com/fig1.png)

We have investigated the structural properties of as-grown and annealed (750 and 800 °C) digital alloy InGaAlAs (λ = 1.3 μm) laser structures by cross-sectional scanning-tunneling microscopy. We show that it is possible to resolve the digital alloy period in the as-grown sample and the 750 °C annealed sample. The 800 °C annealed sample did not show the digital alloy period because of intermixing of the digital alloy. The 750 °C annealed sample showed only slight intermixing. The barrier/well interface roughness for the as-grown and the 750 °C annealed samples was the same. Annealing at 800 °C showed large barrier/well interface roughness and lateral composition modulation due to the phase separation of InGaAs/InAlAs alloys. © 2003 American Institute of Physics.
In Fig. 2(a), we show an STM image of a digitally grown \((\text{InGaAs})_{1-x}(\text{InAlAs})_{x}\) MQW structure. Figure 2(a) was taken at a negative sample bias of \(-1.5\) V, showing the filled states associated with the As sites. The bright regions are the 93.8 (InGaAs)\(_{0.2}\)(InAlAs)\(_{0.8}\) well layers and the dark regions are the 65.6 (InGaAs)\(_{0.4}\)(InAlAs)\(_{0.6}\) barrier layers. The observed contrast between the barrier and the well layers causes an electronic contribution to the tunnel current at low sample bias (\(V_{\text{sample}} < -2\) V). Clearly visible is the period of the digital alloy, which is approximately three atomic bilayers. Figure 2(b) shows a line profile averaged perpendicular to the growth direction. In the well and barrier layers, five respectively, four pairs of digital alloy can be distinguished. This is in agreement with the structure design, as indicated in Fig. 1. Also shown in Fig. 2(b) is the averaged line profile of the MQW region of the 800 °C annealed sample. No digital alloy period can be seen in the line profile, which indicates intermixing of the SPSs occurs after annealing. The intermixing is assumed to cause a blue-shift of the PL spectrum after annealing of these structures.\(^3\)

Generally, contrast in STM images is mostly determined by electronic contributions to the tunnel current. It has been suggested that for the material system InP/GaAs/InAs, it is possible to minimize electronic effects by applying either a high (\(V_{\text{sample}} < 2\) V) positive sample bias or a high negative sample bias (\(V_{\text{sample}} < -2\) V).\(^5,6\)

Figures 3(a) and 3(b) show high-voltage (\(-3\) V), filled-states images of the as-grown and the 800 °C annealed structure. The images have the same height scale, with a maximum of 180 pm. The image of the as-grown sample shows almost no electronic effects, as the barrier and well regions show almost no contrast. The topography is as expected for a homogeneous alloy. The image of the 800 °C annealed structure, however, shows strong lateral corrugation modulation and roughening of the barrier/well interfaces. The roughening of the interfaces can be attributed to barrier/well intermixing due to the interdiffusion of Al and Ga atoms during annealing. The lateral corrugation modulation is caused by a lateral composition modulation due to segregation of indium atoms. It is known that in various material systems, it is possible to have a phase separation into indium-rich and indium-poor phases.\(^7-9\) The bright areas are attributed to indium-rich areas. Note that the filled-states image actually shows the As sites. The indium-rich areas appear bright because the In atoms in the first subsurface layer cause the surface As atoms to protrude out of the surface plane.\(^10\) With
In conclusion, we have shown in this letter that it is possible to resolve the digital alloy period for an as-grown digital alloy InGaAlAs MQW structure by X-STM. Our results show that annealing at 750 °C results in slight intermixing of the SPS, whereas annealing at 800 °C results in complete disappearance of the digital alloy period. Annealing at 750 °C does not affect barrier/well interface roughness. Annealing at 800 °C results in large barrier/well interface roughness and lateral composition modulation due to indium phase separation. These conclusions support the assertions in Ref. 3.

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