

Hitachi Ltd., for their support in this work. They are also grateful to S. Yamashita of the same laboratory for his extensive assistance in diode preparation and scanning microscope measurements.

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Embedded heterostructure epitaxy: A technique for two-dimensional thin-film definition*

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(Received 2 June 1975)

Selective multilayer epitaxial growth of GaAs-Ga_{1-x}Al_xAs through stripe openings in Al₂O₃ mask is reported. The technique results in prismatic layers of GaAs and Ga_{1-x}Al_xAs 'embedded' in each other and leads to controllable uniform structures terminated by crystal faces. The crystal habit (shape) has features which are favorable for fabrication of cw injection lasers, laser arrays, and integrated optics components which require planar definition.

PACS numbers: 68.50., 81.20.F

Multilayer GaAs-GaAlAs thin-film structures grown by epitaxial techniques have been playing an increasingly important role in a variety of electro-optical applications. These include cw injection lasers, distributed-feedback lasers, light-emitting diodes, and a number of electro-optical components for integrated optics. The lateral definition often needed in these applications is achieved by techniques such as proton bombardment,^{1,2} ion etching,² diffusion,^{3,4} chemical etching,^{5,6} chemical etching and regrowth.^{7,8}

In the present paper we report on a new technique for growing multilayer GaAs-Ga_{1-x}Al_xAs through openings in Al₂O₃ masks by liquid-phase epitaxy. Single-layer vapor-phase⁹⁻¹² and liquid-phase^{13,14} epitaxial growth of GaAs and InGaAs through openings in SiO₂ masks have been reported previously. SiO₂ masks are probably not suitable for the growth of GaAlAs because the aluminum in the melt may react with the silicon oxide.¹⁵ We found sputtered Al₂O₃ masks stable and adhering throughout the multilayer growing process. No growth was observed on the oxide away from the window openings.

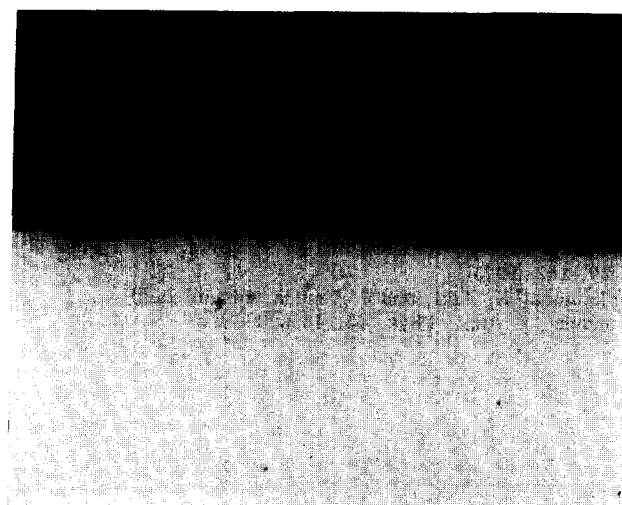
The resulting structure consists of prisms of GaAs, or GaAlAs completely embedded epitaxially in outer crystalline layers of similar material. We thus propose the term embedded epitaxy for this growth technique.

The growth took place on [100]-oriented polished GaAs wafers with (*n* type) Si doping $n = 3 \times 10^{18} \text{ cm}^{-3}$. The wafer's surfaces were sputtered with 3000 Å of Al₂O₃ and were cleaved into rectangles at the two perpendicular

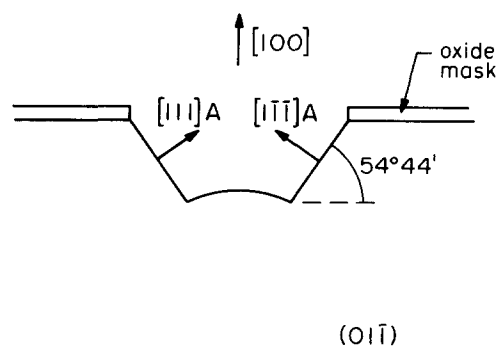
cleavage planes (011). Standard photolithographic techniques were used to define on a wafer photoresist stripe openings of widths 15, 25, and 100 μ on top of the Al₂O₃, and by etching in hot phosphoric acid very smoothly edged stripe openings were formed in the oxide. The stripes were oriented parallel to the cleavage plane (011) or (011) to within ±0.05°.

The wafers were briefly etched in Br₂-methanol and the growth performed by standard multilayer horizontal liquid-phase-epitaxy procedures.¹⁶ The growth temperature was 818 °C and the cooling rate was as slow as 0.1°/min. The cooling cycle was up to an order of magnitude shorter than what is usually used in ordinary epitaxial growth. The first and third layer were Ga_{0.6}Al_{0.4}As [dark layers in Figs. 1(a) and 2(a)]. The second and fourth layers were GaAs [white areas in Figs. 1(a) and 2(a)]. The samples were then cleaved perpendicular to the stripes and the multilayer structure revealed [Figs. 1(a) and 2(a)] using HF:HNO₃:H₂O (1:3:4) staining solution. The multilayer growth technique has the important advantage of indicating the growth history (by revealing the layers). This allowed us to achieve better understanding of the growth pattern in different orientations and conditions, and to improve our control of the growth cycle.

The first noteworthy characteristic is the considerably higher rate of growth through the openings in comparison with unmasked wafers. Furthermore, the thinner the stripe the higher the growth rate. This shows that the amount of growth is determined mainly by the



(a)



(b)

FIG. 1. Cross-sectional view of stripe, perpendicular to $(01\bar{1})$ cleavage plane. (a) Multilayer growth pattern through the stripe. (b) Reconstruction of preferential etching experiment which indicates that the side faces of the pattern of Fig. 1(a) are $(111)A$ faces.

amount of excess supercooled GaAs in the solution. Consequently the growth is faster the smaller the available area. In order to achieve thin layers the cooling rate and the duration of growth must be considerably reduced relative to those in unmasked wafers.

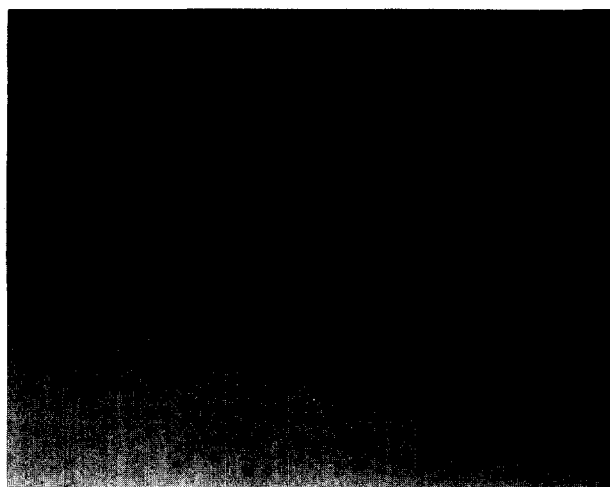
Another important feature is the dependence of the growth pattern on the stripe orientation. In Fig. 1 the stripes are perpendicular to the $(01\bar{1})$ face and the growth overlays the oxide surface. In Fig. 2 the stripes are perpendicular to (011) face and the overlay is minimal. Growth in the directions of the side faces is enhanced relative to the vertical direction in the structure of Fig. 1 while the opposite is true in Fig. 2. An exception to this is the second GaAs layer which in both cases showed thicker growth in the vertical direction. The side faces of the two structures were found in both cases to form an angle of about 55° with the (100) face. This identifies them as (111) surfaces which ideally form an angle of $54^\circ44'$ with the (100) face.

Etching the GaAs through the stripes with a preferential etch (Br_2 -methanol) helps to reveal the sense of orientation of each of the faces of the structures shown in Figs. 1(a) and 2(a). The etch pattern is different in

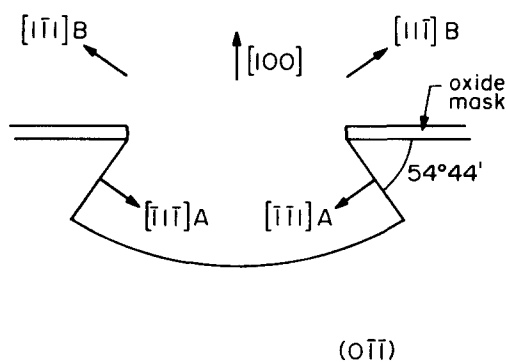
both cases. Since Br_2 -methanol has a higher etching rate for $(111)B$ (As) faces than for $(111)A$ (Ga) faces, it tends to open up channels with $(111)A$ side walls.^{17,18} This identifies the two side faces of the structure in Fig. 1(a) as $(111)A$ (Ga) faces and those of Fig. 2(a) as $(111)B$ (As) faces. One can see clearly from the pictures that the (100) face grows at a slower rate than the $(111)A$ faces (as in Fig. 1) but at a faster rate than $(111)B$ faces (as in Fig. 2). This is in agreement with the result of vapor-phase epitaxy reported by Shaw.¹⁹

It is worth mentioning that the faces of the structures were smooth and highly reflecting and of better quality than those grown on unmasked wafers. (See also discussion in Ref. 10). It is also likely that since the lateral dimensional control is achieved without resort to etching or ion implantation, and terminates with as grown crystal faces, that this procedure leads to a smaller density of nonradiative recombination centers and hence to improved light emission and laser performance.

The authors would like to acknowledge the guidance of Dr. M.B. Panish and S. Sumski on epitaxial crystal



(a)



(b)

FIG. 2. Cross-sectional view of stripe, perpendicular to (011) cleavage plane. (a) Multilayer growth pattern through the stripe. (b) Etching through the stripes indicating that the side faces of the prism of Fig. 2(a) are $(111)B$ faces.

growth, the help of Dr. H. Garvin from Hughes Research Center in sputtering the Al_2O_3 layers, and helpful discussions with Professor J. McCaldin of Caltech.

*Work supported by the Office of Naval Research and by the National Science Foundation Optical Communication Program.

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Laser emission at 1.32 μm from atomic iodine produced by electrical dissociation of $\text{CF}_3\text{I}^\dagger$

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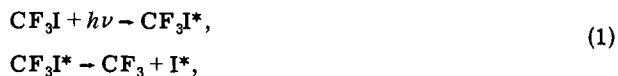
(Received 2 June 1975; in final form 31 July 1975)

Laser emission on the $\text{I}(^2P_{1/2}) \rightarrow \text{I}(^2P_{3/2})$ transition at 1.32 μm has been excited for the first time in an electrical discharge. A mixture of about 1% CF_3I in N_2 was excited in a uv-initiated pulsed discharge between planar electrodes. Iodine atoms in the upper and lower laser levels are generated by dissociative excitation of CF_3I molecules upon electron impact. The lower laser level is depopulated by rapid recombination of the iodine atoms.

PACS numbers: 42.60.C, 82.40.T, 52.80.

In this letter we report the observation of laser emission from the $\text{I}(^2P_{1/2}) \rightarrow \text{I}(^2P_{3/2})$ transition at 1.32 μm excited by the electrical dissociation of CF_3I . These observations are the first reported evidence that a population inversion can be produced on this transition by processes within an electrical discharge.

Investigations of the excitation of the 1.32- μm iodine transition by flash photolysis of CF_3I and other perfluoriodides have been reported extensively in the literature.¹⁻³ The primary process populating the $\text{I}(^2P_{1/2})$ upper state, denoted as I^* , is



where CF_3I^* is the predissociative state corresponding to the optical absorption band in CF_3I at 260 nm. Excitation processes analogous to those found in the iodine photodissociation laser should be produced by inelastic collisions with electrons. In the case of CF_3I , the process populating the upper state would be



Electrons with energies ≥ 4.8 eV would be required to excite the same state as in reaction (1).

The generation of diffuse electrical discharges in CF_3I is complicated by the rapid removal of electrons by dissociative attachment:



The cross section for this reaction is large.⁴ Diffuse electrical discharges in such attaching gases tend to be unstable and there is a tendency to form constricted arcs. We find that this problem can be overcome if the discharge current pulse is terminated before significant amounts of the negative ion are created. For these experiments a pulsed uv-initiated self-sustained discharge between planar electrodes has been employed. Diffuse discharges have been produced in moderate pressures of CF_3I with N_2 and other buffer gases. Laser emission on the 1.32- μm atomic iodine line has been observed over a wide range of conditions in these discharges.

The construction and operating characteristics of the electrical discharge system are similar to those described previously for uv-initiated CO_2 lasers.⁵ The electrodes were fabricated from 304 stainless steel,