

Cathodoluminescence measurement of an orientation dependent aluminum concentration in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ epilayers grown by molecular beam epitaxy on a nonplanar substrate

Michael E. Hoenk, Howard Z. Chen, Amnon Yariv, Hadis Morkoç,^{a)}
and Kerry J. Vahala

Department of Applied Physics, 128-95, California Institute of Technology, Pasadena, California 91125

(Received 16 November 1988; accepted for publication 30 January 1989)

Cathodoluminescence scanning electron microscopy is used to study $\text{Al}_x\text{Ga}_{1-x}\text{As}$ epilayers grown on a nonplanar substrate by molecular beam epitaxy. Grooves parallel to the $[0\bar{1}\bar{1}]$ direction were etched in an undoped GaAs substrate. Growth on such grooves proceeds on particular facet planes. We find that the aluminum concentration in the epilayers is dependent on the facet orientation, changing by as much as 35% from the value in the unpatterned areas. The transition in the aluminum concentration at a boundary between two facets is observed to be very abrupt.

Advances in crystal growth technology have led to increased control of the crystal composition during the growth process. An example of this is the monolayer precision with which the composition of semiconductor crystals can be varied using molecular beam epitaxy (MBE). Since MBE growth on planar substrates affords control of the composition only along the growth axis, additional processing is required to introduce lateral variation of the crystal properties. The use of nonplanar substrates produces lateral variations in epitaxial layers grown by MBE, adding a new dimension to the power of MBE as a tool for device fabrication.¹⁻⁶ For example, growth on nonplanar substrates has been used for the fabrication of buried-heterostructure quantum well lasers in a single growth step,⁵ and it has been proposed as a technique for the fabrication of quantum wire lasers.^{5,6}

Lateral variations in the properties of epilayers grown by MBE on nonplanar substrates occur as a result of an orientation dependence of the surface kinetics during the growth. For example, the growth rate within a given layer varies with position according to the orientation of the surface, and certain dopants (e.g., silicon) appear preferentially as donors on some facets and as acceptors on others.³⁻⁸ In this letter we present cathodoluminescence data on $\text{Al}_x\text{Ga}_{1-x}\text{As}$ epilayers grown on a patterned GaAs substrate. Our data show that the aluminum concentration on some of the facets differs by as much as 35% from the value in the unpatterned areas. The transition between the compositions of two adjacent facets is observed to be very abrupt.

The sample used in this experiment was prepared as follows. Lines $5\ \mu\text{m}$ wide were defined by photolithography on an undoped, high-resistivity GaAs substrate along the $[0\bar{1}\bar{1}]$ direction. A 60 min chemical etch, using a 3:1:40 mixture of $\text{H}_3\text{PO}_4\text{:H}_2\text{O}_2\text{:H}_2\text{O}$, defined grooves in the substrate which were approximately $15\ \mu\text{m}$ wide at the top. The wafer was then cleaned using a series of solvents, none of which acts as etchants for GaAs or AlGaAs. The growth was done in a Riber 2300 molecular beam epitaxy machine. The substrate temperature was $600\ ^\circ\text{C}$. The substrate was rotated at 48 rpm

during the growth to average out any geometrical effects regarding the fluxes of molecules. The structure in the unpatterned areas of the sample consists of a $1\ \mu\text{m}$ GaAs buffer layer, a $1\ \mu\text{m}$ $\text{Al}_{0.248}\text{Ga}_{0.752}\text{As}$ layer, a $100\ \text{\AA}$ GaAs quantum well, and a $0.1\ \mu\text{m}$ $\text{Al}_{0.248}\text{Ga}_{0.752}\text{As}$ cap layer. All layers were undoped.

Cathodoluminescence scanning electron microscopy⁹ of the sample shows a dramatic spatial dependence of the material composition on a nonplanar region of the substrate. Growth on nonplanar regions occurs preferentially along particular facet planes. The composition of the grown layers is found to depend on the orientation of the facets, as illustrated in Fig. 1. This is demonstrated by spectrally resolved cathodoluminescence (SRCL) micrographs taken at a sample temperature of 77 K. The SRCL images are formed by excitation of a small region of the sample with a raster scan of the electron beam.⁹ A monochromator is used as a bandpass filter for the cathodoluminescence. As the center wavelength of the SRCL images is varied from 6600 to 7000 \AA , the various facets exhibit markedly different luminescence features [see Figs. 2 and 3, (a)–(d)]. At a center wavelength of 6600 \AA , facet *c* luminesces strongly, facets *a* and *e* are relatively

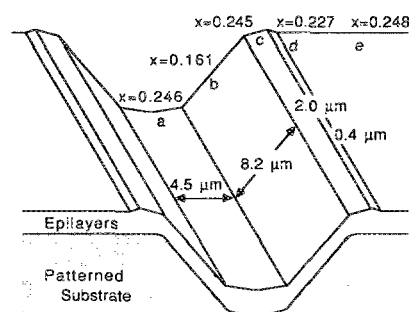


FIG. 1. Diagram of the AlGaAs epilayers grown by molecular beam epitaxy on a nonplanar substrate. The observed aluminum concentrations on the various facets are marked on the diagram. The grooves are symmetric about the center, both with regard to their morphology and the composition of the epilayers.

^{a)} On leave from the University of Illinois, Coordinated Science Laboratory, Urbana, IL 61801.

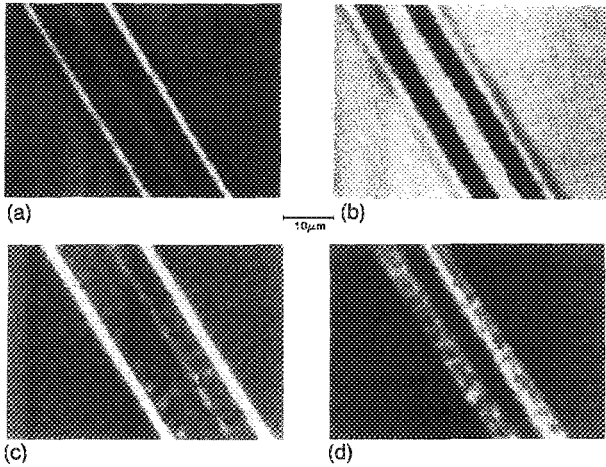


FIG. 2. Spectrally resolved cathodoluminescence micrographs of a groove in the surface. Growth in the groove takes place on well-defined facets (see Fig. 1). The different micrographs represent luminescence images of the sample taken at particular wavelengths. (a) 6600 Å: Facet *c* appears bright, and facets *a* and *e* are dim. (b) 6700 Å: Facets *a*, *c*, and *e* are bright. (c) 6800 Å: Facet *d* is bright. (d) 7000 Å: Facet *b* is bright.

dim, and facets *b* and *d* are dark [see Figs. 2(a) and 3(a)]. In Figs. 2(b) and 3(b), the center wavelength is 6700 Å, and facets *a*, *c*, and *e* are luminescing strongly, while facet *b* is dark and facet *d* is relatively dim. With the center wavelength at 6800 Å, facet *d* luminesces quite strongly, and all other facets are relatively dark [see Figs. 2(c) and 3(c)]. At 7000 Å, the luminescence is dominated by facet *b*, and all other facets are dark [see Figs. 2(d) and 3(d)]. The observed contrast in the SRCL images is due to different amplitudes, widths, and positions of the peaks in the luminescence spectra emitted from the various facets (see Fig. 4). In particular, the unpatterned region (facet *e*) has an $\text{Al}_x\text{Ga}_{1-x}\text{As}$ luminescence peak at 6672 Å, corresponding to an aluminum concentration of $x = 0.248$ (see Fig. 4, dotted line).¹⁰ The bottom of the groove, facet *a*, has a luminescence peak at 6682 Å, which corresponds to an aluminum concentration of $x = 0.246$ (see Fig. 4, solid line). The top of the groove, facet *c*, has a luminescence peak at

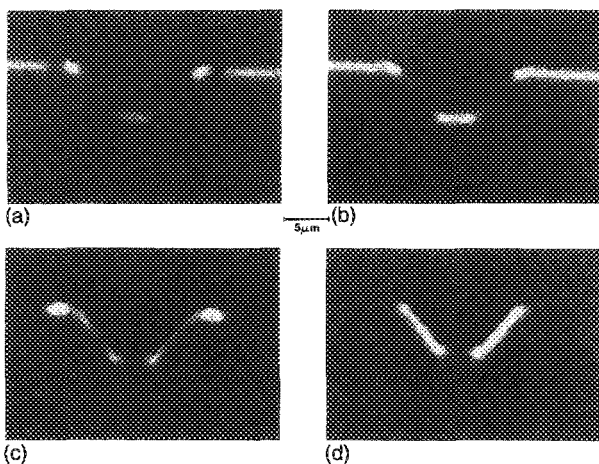


FIG. 3. Spectrally resolved cathodoluminescence micrographs of a cross section of a groove in the surface. Growth in the groove takes place on well-defined facets (see Fig. 1). The different micrographs represent luminescence images of the sample taken at particular wavelengths. (a) 6600 Å: Facet *c* appears bright, and facets *a* and *e* are dim. (b) 6700 Å: Facets *a*, *c*, and *e* are bright. (c) 6800 Å: Facet *d* is bright. (d) 7000 Å: Facet *b* is bright.

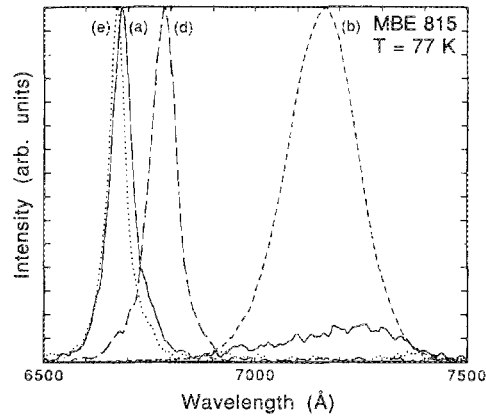


FIG. 4. Cathodoluminescence spectra measured using spot excitation of a groove in the sample (see Fig. 1 for facet designation, *a* through *e*). The vertical scales of the spectra are different. Solid line: spectrum from facet *a*, the bottom of the groove. Short dashes: spectrum from facet *b*, the side of the groove. Long dashes: spectrum from facet *d*. Dotted line: spectrum from the unpatterned area. The spectrum from facet *c*, not shown, resembles the spectrum from facet *a*, except that the peak near 6700 Å is broader.

6687 Å and an aluminum concentration of $x = 0.245$. Facet *d*, the narrow facet adjacent to the unpatterned region, has a luminescence peak at 6780 Å, corresponding to an aluminum concentration of $x = 0.227$ (see Fig. 4, long dashes). The side of the groove, facet *b*, has its luminescence peak at 7160 Å, corresponding to an aluminum fraction of $x = 0.161$ (see Fig. 4, short dashes). Thus, from the bottom of the groove to the sidewall there is a change in the aluminum concentration of 35%.

In order to determine the abruptness of the change in the aluminum concentration between the bottom of the groove, facet *a*, and the side of the groove, facet *b*, we took a series of spectra at various points on these facets. It should be mentioned that the spatial resolution of cathodoluminescence images and spectra is limited by the size of the interaction volume in which electron-hole pairs are created, and by diffusion of carriers prior to recombination. The interaction volume is roughly 0.5 μm in diameter for a 10 keV electron beam, and the diffusion length is on the order of a micron. Thus, the peaks in the spectra correspond to transitions which take place in a volume of material with a radius on the order of a micron. As we move from the center of the groove to the border between the bottom and the side, the peak at 6682 Å gradually shifts to 6710 Å, and a peak at 7050 Å appears (see Fig. 5). As we move from there to the center of the sidewall, the peak at 6710 Å disappears, and the peak at 7050 Å shifts to 7160 Å. The border between the bottom of the groove and the sidewall is relatively bright in the SRCL image in Fig. 3(c). This bright area corresponds to a superposition of the tails of the peaks at 6710 and 7050 Å. At every point sampled, the two peaks were distinct from one another, and no peaks at an intermediate wavelength were observed. This indicates that the transition in the aluminum concentration at the boundary between the facets is very abrupt. Because of the finite resolution of cathodoluminescence scanning electron microscopy, we cannot measure precisely the length scale on which the transition occurs, but it is possible that a lateral heterojunction exists at the boundary between facets.

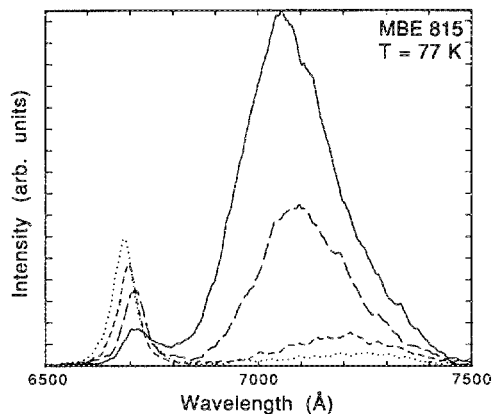


FIG. 5. Cathodoluminescence spectra measured using spot excitation at various points around the border between the bottom of the groove (facet *a*) and the side of the groove (facet *b*). See Fig. 1 for a diagram of the facets. The two peaks remain distinct through the transition from facet *a* to facet *b*, indicating that the aluminum concentration changes abruptly at the facet boundary. Dotted line: spot excitation of the center of facet *a*. Short dashes: excitation spot moved roughly $0.7 \mu\text{m}$ from the center of facet *a*. Long dashes: spot moved roughly $1.4 \mu\text{m}$ from the center of facet *a*. Solid line: excitation of the border between facets *a* and *b*.

We found one place in our sample which, through a flaw in the photolithography, had a bridge across one of the grooves. The contour of the bridge was analogous to a "groove" etched in the $[01\bar{1}]$ direction. In this region the facet which developed on the side of the groove exhibited luminescence peaked at about 7270 \AA , corresponding to an aluminum concentration of $x = 0.143$. This represents a 42% change from the unpatterned area, which is even larger than the result for the $[01\bar{1}]$ groove.

In order to make sure that the presence of the quantum well did not alter the spectra of the AlGaAs layers, approximately $0.25 \mu\text{m}$ of material from the top of the wafer was removed with a two min etch in the 3:1:40 solution. Cathodoluminescence spectra verified that the quantum well was absent in the etched sample, and that the AlGaAs luminescence spectra were unchanged.

Deviation of the aluminum concentration from the value in the unpatterned region of the sample occurs because of a difference in the kinetics of growth for aluminum and gallium atoms.^{11,12} There are two mechanisms discussed in the literature for this difference to cause a variation in the local composition.¹³ First, aluminum and gallium have different sticking coefficients, defined on infinite surfaces as the ratio of the number of atoms incorporated into the surface to the incident flux. Second, aluminum and gallium have different surface diffusion lengths, so that near a facet boundary, the population of surface atoms may be altered by transfer of atoms to an adjacent facet. Growth of our sample was done at low temperature, 600°C . For MBE growth on the $[100]$ plane at this temperature, the sticking coefficients of aluminum and gallium are close to unity,¹¹ and their diffusion lengths are on the order of a few hundred angstroms or less.^{14,15} Since the combination of unity sticking coefficients and short diffusion lengths would lead to a uniform aluminum concentration in the grooves, these parameters must be strongly dependent on the facet orientation in order to account for our cathodoluminescence observations. In partic-

ular, the abruptness of the transition between the aluminum concentrations on facets located at the bottom and the side of the groove indicates that the behavior of aluminum and gallium is quite different on the two facets.

The aluminum concentrations on the various facets are expected to depend on the growth parameters. The growth rate of epilayers on different facets has been found to be strongly dependent on the growth conditions, such as substrate temperature and As/Ga flux ratio.^{3,4} Since there is a connection between the growth rate and the sticking coefficients of the group III elements,⁶ the aluminum concentration on a given facet should also depend on the growth parameters. This raises the possibility that further optimization of the structure may be done using the growth conditions as a patterning tool. For example, if it is found that certain growth conditions affect the aluminum concentration on the facets, changing these conditions during the growth could produce an interesting three-dimensional variation of the aluminum concentration, with possible applications toward making devices.

In conclusion, we have observed a strong dependence of the composition of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ epitaxial layers grown by MBE on a nonplanar substrate using cathodoluminescence scanning electron microscopy. The aluminum concentration was observed to change from $x = 0.246$ on the bottom of the groove to $x = 0.161$ on the side of the groove. The transition in the aluminum concentration at a boundary between two facets is observed to be very abrupt. While the spatial resolution of cathodoluminescence scanning electron microscopy is not sufficient to determine the length scale on which the transition takes place, it is possible that a lateral heterojunction exists at the boundary between facets.

The authors would like to acknowledge Dhruves Biswas for his contribution of data on the thickness of GaAs layers grown on a nonplanar substrate. This work was supported by the Office of Naval Research.

¹W. T. Tsang and A. Y. Cho, *Appl. Phys. Lett.* **30**, 293 (1977).

²Seiichi Nagata, Tsuneo Tanaka, and Masakazu Fukai, *Appl. Phys. Lett.* **30**, 503 (1977).

³John Stephen Smith, Pamela L. Derry, Shlomo Margalit, and Amnon Yariv, *Appl. Phys. Lett.* **47**, 712 (1985).

⁴John Stephen Smith, Ph.D. thesis, Caltech, Dept. of Appl. Phys., 1986.

⁵E. Kapon, J. P. Harbison, C. P. Yun, and N. G. Stoffel, *Appl. Phys. Lett.* **52**, 607 (1988).

⁶E. Kapon, M. C. Tamargo, and D. M. Hwang, *Appl. Phys. Lett.* **50**, 347 (1987); E. Kapon, D. M. Hwang, R. Bhat, and M. C. Tamargo, *Superlatt. Microstruct.* **4**, 297 (1988).

⁷W. I. Wang, E. E. Mendez, T. S. Kuan, and L. Esaki, *Appl. Phys. Lett.* **47**, 826 (1985).

⁸D. L. Miller, *Appl. Phys. Lett.* **47**, 1309 (1985).

⁹Michael E. Hoenk and Kerry J. Vahala, *Rev. Sci. Instrum.* **60**, 226 (1989).

¹⁰F. F. Kuech, D. J. Wolford, R. Potemski, J. A. Bradley, K. H. Kelleher, D. Yan, J. Paul Farrell, P. M. S. Lesser, and F. H. Poliak, *Appl. Phys. Lett.* **51**, 505 (1987).

¹¹R. Fischer, J. Klem, T. J. Drummond, R. E. Thorne, W. Kopp, H. Morokoc, and A. Y. Cho, *J. Appl. Phys.* **54**, 2508 (1983).

¹²S. B. Ogale, M. Thomsen, and A. Madhukar, *Appl. Phys. Lett.* **52**, 723 (1988).

¹³Mitsuru Ohtsuka and Seiichi Miyazawa, *J. Appl. Phys. Lett.* **64**, 3522 (1988).

¹⁴S. Nagata and T. Tanaka, *J. Appl. Phys.* **48**, 940 (1977).

¹⁵J. H. Neave, P. J. Dobson, B. A. Joyce, and Jing Zhang, *Appl. Phys. Lett.* **47**, 100 (1985).