CONF-931108--115

LBL-35470
UC-411

Center for Advanced Materials

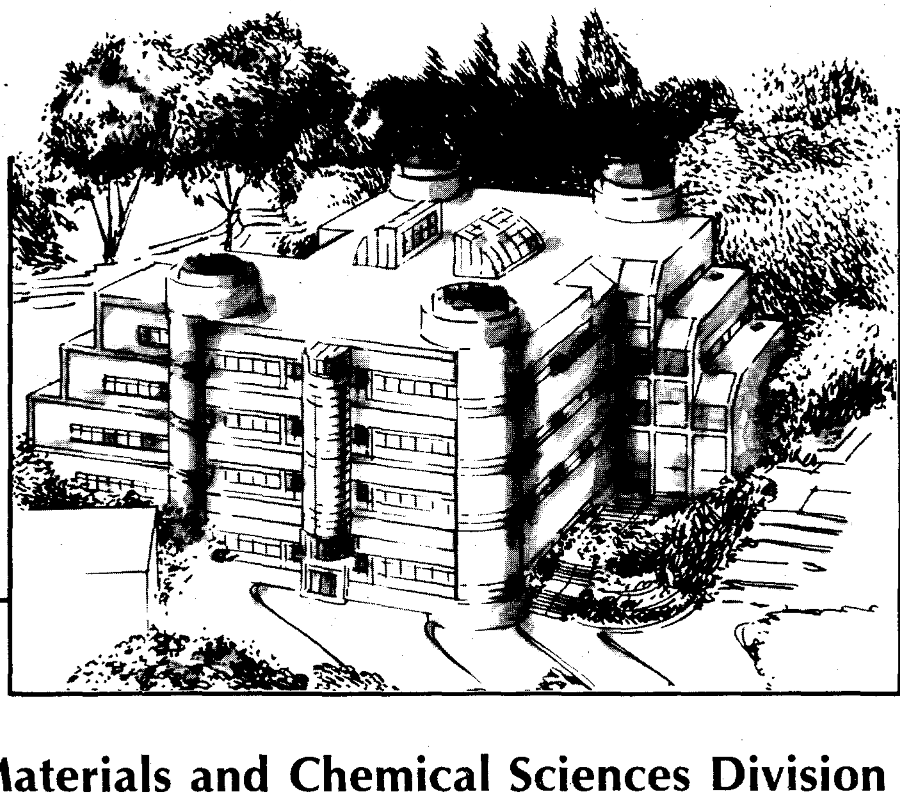
CAM

Presented at the 1993 Fall Meeting of the Materials Research Society, Boston, MA, November 29–December 2, 1993, and to be published in the Proceedings

Atomic Scale Interface Structure of $In_{0.2}Ga_{0.8}As/GaAs$ Strained Layers Studied by Cross-Sectional Scanning Tunneling Microscopy

J.F. Zheng, M.B. Salmeron, and E.R. Weber

November 1993



Materials and Chemical Sciences Division

Lawrence Berkeley Laboratory • University of California

ONE CYCLOTRON ROAD, BERKELEY, CA 94720 • (415) 486-4755

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

LBL-35470
UC-411

Atomic Scale Interface Structure of $In_{0.2}Ga_{0.8}As/GaAs$ Strained Layers Studied by Cross-Sectional Scanning Tunneling Microscopy

J.F. Zheng,[1,2] M.B. Salmeron,[2] and E.R. Weber[1,2]

[1]Department of Materials Science and Mineral Engineering
University of California, Berkeley

and

[2]Materials Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

November 1993

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

ATOMIC SCALE INTERFACE STRUCTURE OF $In_{0.2}Ga_{0.8}As$ /GaAs STRAINED LAYERS STUDIED BY CROSS-SECTIONAL SCANNING TUNNELING MICROSCOPY

J. F. Zheng[1,2], M. B. Salmeron[2] and E. R. Weber[1,2]

[1]Department of Materials Science and Mineral Engineering
University of California at Berkeley, Berkeley, CA 94720

[2]Materials Sciences Division, Lawrence Berkeley Laboratory
University of California, Berkeley, CA 94720

ABSTRACT

A molecular beam epitaxy-grown $In_{0.2}Ga_{0.8}As$/GaAs strained layer structure has been studied by scanning tunneling microscopy in cross-section on the (110) cleavage plane perpendicular to [001] the growth direction. Individual indium atoms were differentially imaged in the group III sublattice, allowing a direct observation of the interface roughness due to the indium compositional fluctuation. In the $In_{0.2}Ga_{0.8}As$ layers, Indium atoms are found in clusters preferentially along the growth direction with each cluster containing 2-3 indium atoms. Indium segregation induced asymmetrical interface broadening is studied on an atomic scale. The interface of $In_{0.2}Ga_{0.8}As$ grown on GaAs is sharp within 2-4 atomic layers. The interface of GaAs grown on $In_{0.2}Ga_{0.8}As$ is found to be broadened to about 5-10 atomic layers. The atomic scale fluctuation due to indium distribution is about 20 Å along the interface in this case. We conclude that clustering and segregation are the main reason for the $In_{0.2}Ga_{0.8}As$/GaAs interface roughness.

INTRODUCTION

InGaAs/GaAs strained-layer heterostructures have received increasing attention in recent years because of their potential application in both optoelectronics and field effect transistors. These applications make use of the narrow band gap of InGaAs for infrared optoelectronic devices and the small electron effective mass for high frequency transistors. However, interface quality has been a crucial issue in the use of this heterostructure system. Due to lattice mismatch, high quality InGaAs layer can only be prepared by the controlled pseudomorphic growth of a layer with thickness below the critical value,[1] so that a strained-layer structure is formed without misfit dislocations. The interface quality is influenced by the indium segregation during the growth and is subject to active studies.[2-8]

Atomic scale studies of III/V semiconductor heterostructures are traditionally performed by transmission electron microscopy (TEM).[9] However, the TEM images are formed by 10-100 layers of atoms essentially averaging the information on an atomic level. In addition, TEM does not have chemical sensitivity to differentiate individual atoms in a non-periodic arrangement in the lattice. Photoluminescence (PL) studies of semiconductor quantum well structure interface roughness is limited in resolution by the exciton diameter (~100Å).[10] Scanning tunneling microscopy (STM) is a powerful technique for the study of localized geometric and electronic structures of surfaces.[11] STM

Proc. 1993 Fall Meeting of the Materials Research Society, Boston, MA, November 29-December 2, 1993. **Defect-Interface Interactions Symposium,** eds. E.P. Kvam, A.H. King, M.J. Mills, T.D. Sands and others, pp. 111-116. Materials Research Society, Pittsburgh, 1994.

has shown the capability to distinguish different atomic species via their local electronic structure.[12-13] A recently developed approach applies STM in cross-section, allowing the study of III-V heterostructures with atomic resolution.[14-16]

Here we report the first direct observation of the spatial distribution of individual indium atoms in GaAs/$In_{0.2}Ga_{0.8}As$/GaAs strained layers. Indium is found to cluster along the growth direction within the $In_{0.2}Ga_{0.8}As$. Indium segregation at the GaAs/$In_{0.2}Ga_{0.8}As$/GaAs interfaces and the resulting asymmetrical broadening is also studied on an atomic scale. We propose that clustering and segregation are the main reason for the roughness of GaAs/$In_{0.2}Ga_{0.8}As$/GaAs interfaces.

EXPERIMENTAL

The studies were conducted using STM in cross-section in an ultra-high-vacuum (UHV) environment (8×10^{-11} Torr). The sample is a multiple quantum well structure with three undoped 80Å-thick $In_{0.2}Ga_{0.8}As$ layers as quantum wells separated by two undoped 100Å-thick GaAs layers as barriers. The 80Å $In_{0.2}Ga_{0.8}As$ layer is below the critical thickness so that no dislocations are present and the structure is strained. [1] The sample was grown by MBE at 540°C on [001] oriented n+ GaAs substrate. It is exposed for STM studies by cleavage in UHV along the (110) plane, perpendicular to the [001] growth direction. The STM system used for this study is homemade and has been described previously.[16] The STM tips are electrochemically etched Pt-Rh wires (0.25 mm in diameter), and all images were taken in a constant current mode.

RESULTS AND DISCUSSION

1. Images of individual indium atoms

First we show a 2000Å × 2000Å STM image with the strained-layer GaAs/$In_{0.2}Ga_{0.8}As$/GaAs MQW's viewed in the cross-sectionally cleaved (110) surface (Fig. 1). The three 80Å-thick $In_{0.2}Ga_{0.8}As$ wells are imaged as the brighter bands running from the bottom left to top right and are separated by two slightly darker GaAs layers. The irregular black and white bands from bottom right to the top left are single atomic steps created by the cleavage.

Fig. 1. 2000Å × 2000Å STM image of the cleaved (110) surface. The image is acquired with a tunneling current of 0.5 nA at sample bias of -2.0 V. Three $In_{0.2}Ga_{0.8}As$ multiple quantum wells appear as white bands running from bottom left to top right, superimposed on the atomic steps created by the cleavage.

Zooming in the region enclosed by a box in Fig. 1, we see a 150Å × 120Å STM image with atomic resolution (Fig. 2). It shows the GaAs on InGaAs and the InGaAs on GaAs interfaces. The image is taken at positive sample bias, corresponding to electrons tunneling from the tip to the empty states of the sample surface. Since empty states are located preferentially at the group III (cation) sites, the atoms imaged in the GaAs region are gallium and those in the $In_{0.2}Ga_{0.8}As$ region are gallium or indium. In this image, most important is the presence of bright atoms which are ascribed to Indium atoms at the group III lattice sites as discussed below.

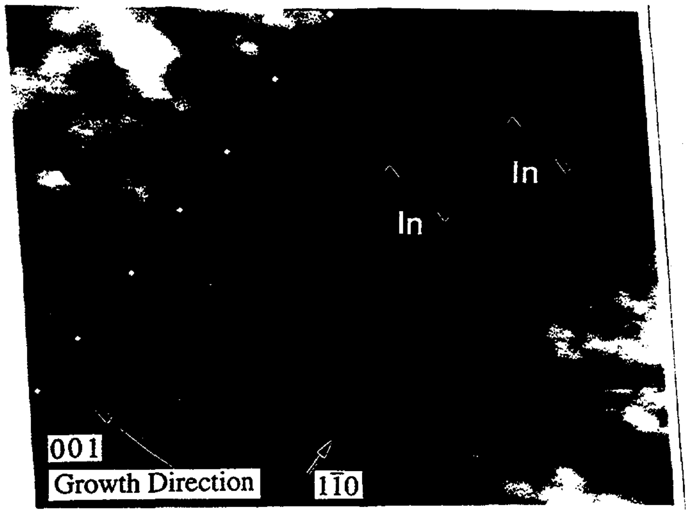


Fig. 2. 150Å × 120Å STM image from obtained the boxed area in Fig. 1. The nominal interface of $In_{0.2}Ga_{0.8}As$ grown on GaAs is indicated by the white dots. The image was acquired with a tunneling current of 0.5 nA at sample bias of +2.0 V. A few indium atoms in clusters are indicated by arrows.

Fig. 3 is an expanded empty state STM image of an area inside $In_{0.2}Ga_{0.8}As$ layer. In this image, the observed individual brighter atoms have higher corrugation (~0.3 Å higher, as seen in Fig. 3b). We assign these brighter atoms to indium, based on the following arguments. First, the fraction of these brighter atoms is constant in all STM images and accounts for 20 ± 5% of all the group III sublattice sites in the images, which agrees well with the nominal concentration of 20% In. Second, since indium is associated with empty states with lower energy than gallium, the tunneling probability is larger when the tip is located at the indium site rather than at the gallium site (Fig. 3c). The higher corrugation of indium is the result of this electronic effect.

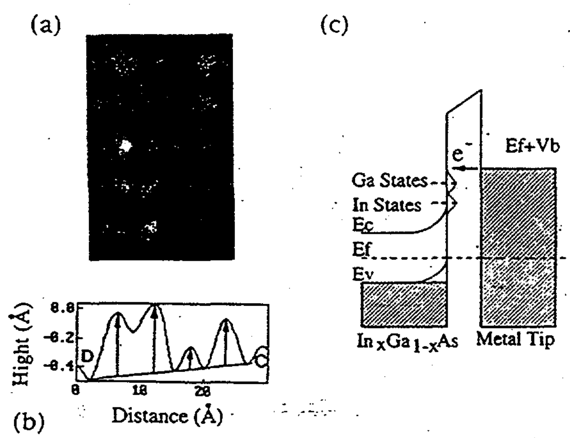


Fig. 3 . (a) Expanded STM image of the empty states in the $In_{0.2}Ga_{0.8}As$ region, showing brighter atoms which are attributed to indium. The image was acquired with a tunneling current of 0.5 nA at a sample bias of +2.0V. (b) Line profile along the arrows at C and D shows the variation in corrugation from the brighter to dimmer atoms. (c) Energy band diagram illustrating the electron tunneling process. The tip-induced band bending at the semiconductor surface is considered. The lower energy of the In-states explains the higher tunneling probabilities and their bright appearance.

2. Indium clustering in $In_{0.2}Ga_{0.8}As$ alloy

Close inspection of many images reveals that the ternary $In_{0.2}Ga_{0.8}As$ alloy region tends to have 2-3 grouped indium atoms that are seen on the (110) plane. We ascribe this to a clustering of indium atoms. A few of these are indicated by arrows in Fig. 2. Statistically, 90% of the clusters contain 2-3 indium atoms. We found that the indium atoms are aligned preferentially along the [001] growth direction, as seen by the chains of brighter atoms in Fig. 2. We disregarded the possibility of plate-like clustering on the (110) planes (perpendicular to the surface) that are imaged edge-on. If it was plate-like on the (110) plane, this should also be seen on the (110) cleavage plane, because it is identical to (110) by symmetry. Thus the clustering was concluded to be preferentially chain-like along the [001] growth direction. In the GaAs region near the $In_{0.2}Ga_{0.8}As$ layer, some slightly bright atoms are due to clusters of indium located in the second layer below the cleavage surface.

We suggest that the observed clustering is the result of local strain. In substitutional ternary III/V semiconductors with two group III elements, there is the possibility of clustering just as a result of random arrangement when the percentage of the substitutional group III elements is large enough. However, random clustering should lead to a wide distribution of cluster sizes.[17] In our results, random clustering is very unlikely since 90% of the clusters contain 2-3 indium atoms. We propose the following explanation for the strain induced clustering (see Fig. 4). During the MBE growth of $In_{0.2}Ga_{0.8}As$ on GaAs, incorporation of indium into the group III lattice will cause strain because its size is larger than that of the gallium. Indium atoms tend to segregate on the growing surface and lead to a high concentration of surface indium adatoms.[8] Near equilibrium, indium atoms can be incorporated into the lattice by kinetic freezing.[2] Once an indium atom is at a group III site on the top growing layer, strain is built up locally. The strain is mostly in the horizontal (001) growth plane because the vertical strain is relaxed due to the free surface. This strain tends to expand the lattice locally, and thus might favor the incorporation of a second indium atom at the site on top of the first one in the following growing layers. However, this process is not likely to form long columns with more indium atoms because the local strain along the clustering direction will prevent the incorporation of additional indium atoms. At this point, the conditions become more favorable for indium incorporation at a new site. In support of this argument, it was found that clusters containing more than three indium atoms occur less frequently.

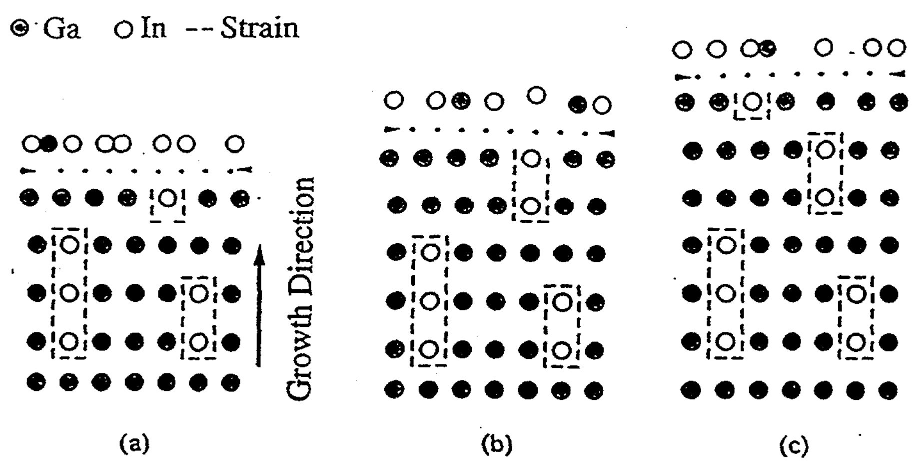


Fig. 4. A schematic model showing the suggested growth process: indium incorporation in group III lattice. The view is on the (110) plane. Arsenic atoms are not shown. Open circles are indium atoms and filled circles refer to gallium atoms. (a) An indium atom is incorporated into the top growing layer. (b) This indium atom expands the lattice horizontally, making the incorporation of another indium on the top site in the next growing layer favorable. (c) When an indium is on top of the other in a solid phase, strain along the growth direction starts to build up. Thus additional indium atoms get more favorable incorporation in a new site.

3. Asymmetrical interface broadening

In Fig. 2, we see directly that the interface of $In_{0.2}Ga_{0.8}As$ grown on GaAs is quite abrupt within the first 2-4 atomic layers. However, the interface of the GaAs grown on $In_{0.2}Ga_{0.8}As$ is diffuse within 5-10 layers, with some indium atoms incorporated deep in the GaAs layer. Fig. 5 shows clearly the asymmetric broadening of the interfaces.

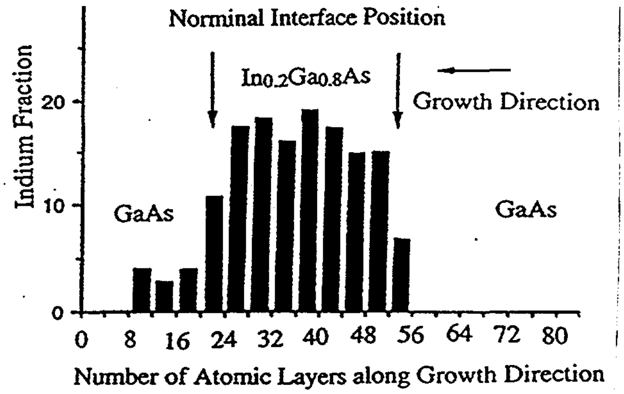


Fig. 5. Distribution of indium atoms across the $GaAs/In_{0.2}Ga_{0.8}As/GaAs$ heterostructure. The vertical bars are calculated by counting the fraction of indium atoms in rows of group III atoms along the (110) direction. The total number of atoms for each bar is about 100. The position of the zero (0) layer is selected arbitrarily.

During MBE growth of InGaAs, excess indium tends to segregate on the growing surface.[8] In the growth of the first few atomic layers of InGaAs when the top growth surface is not uniformly rich with segregated indium, incorporation of indium is inhomogeneous. Thus the $In_{0.2}Ga_{0.8}As$ on GaAs interface is wavy. After several layers, when sufficient segregated indium exist on the growing surface, the $In_{0.2}Ga_{0.8}As$ will grow with more uniform indium incorporation. At the GaAs on $In_{0.2}Ga_{0.8}As$ interface, on the other hand, the excess indium on the growing surface is incorporated into 5-10 layers, resulting in the observed diffused interface. It is interesting to notice that the indium atoms incorporated deep inside the GaAs region also form indium clusters containing ~2 indiums and are oriented along the growth direction, similar to those inside the $In_{0.2}Ga_{0.8}As$. The discussion above allows us to conclude that clustering and segregation are the main reason for interface roughness in this system.

The asymmetrical interface broadening observed here directly is consistent with previous studies by photoelectron spectroscopy[7] and by TEM chemical lattice image studies of InGaAs/AlGaAs heterostructure.[18] The interface roughness fluctuates along the interface with a length scale of about ~20 Å, as directly seen here in Fig. 2. While photoluminescence (PL) indicates flat interfaces of $GaAs/In_{0.2}Ga_{0.8}As/GaAs$ heterostructures,[19] PL does not probe fluctuations smaller than the confined exciton diameter (~100 Å).

CONCLUSIONS

In summary, we have used cross-sectional STM to study atomic scale interface structure of strained-layer $In_{0.2}Ga_{0.8}As/GaAs$ multiple quantum wells. STM is capable of directly imaging the individual indium distribution in $In_{0.2}Ga_{0.8}As$ on an atomic scale. We find that indium clusters preferentially in $In_{0.2}Ga_{0.8}As$ along the growth direction and the clusters contain 2-3 indium atoms. We attribute the clustering to the minimization of local strain during growth. In addition, segregation-induced asymmetrical interface broadening was studied on an atomic scale. The interface of $In_{0.2}Ga_{0.8}As/GaAs$ is abrupt within 2-4 atomic layers and that of $GaAs/In_{0.2}Ga_{0.8}As$ is broad to ~5-10 atomic layers. We suggest that the roughness of the $In_{0.2}Ga_{0.8}As/GaAs$ interface is the result of indium clustering and segregation.

ACKNOWLEDGMENTS

We thank Z. Liliental-Weber, J. Washburn, D.F. Ogletree and J.D. Joannopoulos for fruitful discussions. This work was supported by the Lawrence Berkeley Laboratory through the Director, Office of Energy Research, Office of Basic Energy Science, Materials Science Division, U.S. Department of Energy under contract No. DE-AC03-76SF00098.

REFERENCES

1. J.W. Matthews and A.E. Blakeslee, J. Crystal Growth **27**, 118 (1974).
2. J.M. Gerard and J.Y. Marzin, Phys. Rev. B **45**, 6313 (1992).
3. B. Jojai and P.W. Yu, Phys. Rev. B **41**, 12650 (1990).
4. R.L.S. Devine and W. T. Moore, J. Appl. Phys. **62**, 3999 (1987).
5. P.B. Kirby, J.A. Constable and R. S. Smith, Phys. Rev. B **40**, 3013 (1989).
6. K. Muraki, S. Fukatsu, Y. Shiraki and R. Ito, J. Crystal Growth **127**, 546 (1993).
7. J. M. Larive, J. Nagle, J. P. Landesman, X. Marcadet, C. Mottet and P. Bois, J. Vac. Sci. Technol. B **11**, 1413 (1993).
8. J.M. Moison, F. Houzay, F. Barthe, J.M. Gerard, B. Jusserand, J. Massies and F.S. Turco-Sandroff, J. Crystal Growth **111**, 141 (1991).
9. A. Ourmazd, D.W. Taylor, J. Curningham and C.W. Tu, Phys. Rev. Lett. **62**, 933 (1989).
10. C.A. Warwick, W.Y. Jan, A. Ourmazd and T.D. Harris, Appl. Phys. Lett. **56**, 2666 (1990).
11. R. M. Feenstra, J.A.Stroscio, J. Tersoff and A.P. Fein, Phys. Rev. Lett. **58**, 1192 (1987).
12. R.J. Hamers, J. Vac. Sci. Technol. B **6**, 1462 (1988).
13. J. F. Zheng, X. Liu, E.R. Weber, D.F. Ogletree and M.B. Salmeron, J. Vac. Sci. Technol. B **12** (3), 2104 (1994).
14. O. Albrektsen and H. Salemink, J. Vac. Sci. Technol. B **9**, 779 (1991).
15. H. Salemink and O. Albrektsen, Phys. Rev. B **47**, 16044 (1993).
16. J.F. Zheng, J.D. Walker, D.F. Ogletree, M.B. Salmeron and E.R. Weber, J. Vac. Sci. Technol. B **12** (3), 2100 (1994).
17. N. Holonyak, Jr., W.D. Laidig, M.D. Camras, H. Morkoç, T.J. Drumond, K. Hess and M.S. Burroughs, J. Appl. Phys. **52**, 7201 (1981).
18. J. Kim, J.J. Alwen, D.V. Forbes, J.J. Coleman, I.M. Robertson, C.M. Wayman, F.H. Baumann, M.Bode, Y. Kim and A. Ourmazd. Appl. Phys. Lett. **61**, 28 (1992).
19. K. Muraki, S. Fukatsu, Y. Shiraki and R. Ito, Surf. Sci. **267**, 107 (1992).