Growth of Aluminum Film on GaAs(110) and GaP(110) observed by LEED

By Naoyuki MIYATA*

(Received July 15, 1977)

Abstract

Aluminum films deposited on air-cleaved and heat-cleaned surfaces of GaAs (110) and GaP (110) were in situ observed by LEED. The substrate temperature lay between 20°C and 200°C. In systems Al-GaAs the first appearance of the LEED pattern was streaks extending along [001] direction of the substrate in the substrate temperature range from 150°C to 200°C, the spacing between streaks corresponding to the normal Al–Al interatomic distance. After increasing the thickness {111} facets were formed. The bases of the facets were given to be (112) and (341), (431), the orientation of the former being Al(112)//GaAs(110) and Al[$\bar{1}$10] //GaAs[$\bar{1}$10]. At low temperature, only (111) facet was formed, the bases of the facet was drived to be (25 14 1) and (14 25 1). In systems Al–GaP, (111) facet was formed, the base of the facet was drived to be (341) and (431).

1. Introduction

Studies of the epitaxial growth of some metallic films on (110) cleaved surfaces of zincblende type crystals (Au, Cu, Ni on ZnS1) and Ag on GaP2), GaAs3)) have revealed a few degree of inclination of the (110) film plane to the substrate (110) surface. The latter carried out under U. H. V. showed the presence of facet of facet systems, while the former made under not very clean vacuum indicated no such features. Moreover, our works made on systems of Ag, Au on GaAs3)4) and Au on GaP5) have shown that the facet formation in these films were asymmetric with respect to the substrate [$\bar{1}$10] axis. The asymmetric structure stated above strongly suggests the effect of the asymmetric atomic arrangement in the (110) surface of zincblende type crystal on the film growth. Remarkable differences in the film structure have been observed between Ag and Au on the same substrate. Thus we intended to extend our study to systems of metallic films on cleaved surfaces GaAs and GaP which have zincblende structure, in order to study the mechanism of film growth on this sort of the substrate. In the present study results obtained for systems Al–GaAs and Al–GaP will be given.

2. Experimental

A post-acceleration display type LEED optics with two spherical grids was used. Aluminum of 99.99% purity was evaporated from a tungsten helical basket heater at a

---

* Department of Electronics

rate ranging from a few tens Å/min. to several tens Å/min.. The thickness of the film or deposition rate was estimated indirectly by measuring the thickness of an aluminum film deposited on a small glass plate which had been placed midway between the source and the substrate so as to receive a part of the evaporated vapor. LEED observations were made intermittently during the deposition to follow the growth process, the maximum thickness examined being about 500 Å. The substrates used in this study were undoped GaAs and GaP crystals cleaved in air. The specimen easily produced a well defined, low background, (1 × 1) LEED patterns as shown in Fig. 1 for GaAs and Fig. 2 for GaP after heating up to about 600°C within a few minutes in the vacuum of 10^{-9} Torr. The substrate was held in a folded thin tantalum foil heater attached to the specimen manupulator, and the temperature of the substrate was measured by pressing a fine chromel-alumel thermocouple to the back side of the crystal. The absolute [001] direction in the (110) surface was determined by cleaving the crystal after a set of (111) and $(\bar{1}\bar{1}\bar{1})$ planes had been found out by etching method6). The intensity measurement was carried out by a spot photometer. Intensity curves of (01) and $(0\bar{1})$ spots are shown in Fig. 3(a) for GaAs and Fig. 3(b) for GaP, the former being quite similar to MacRae's curve7). It's evident that the remarkable intensity difference between (01) and $(0\bar{1})$ at some voltage range can be used to determine the absolute [001] direction.

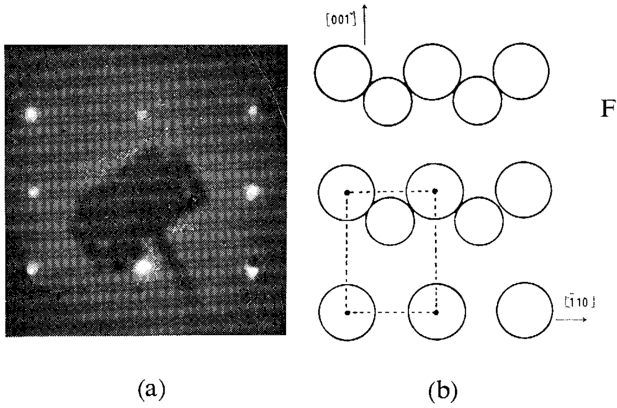


Fig. 1 A LEED pattern obtained from clean GaAs (110) surface.
(a) Photographed at 32 eV.
(b) Atomic arrangement of a unit cell of the GaAs (110) surface.

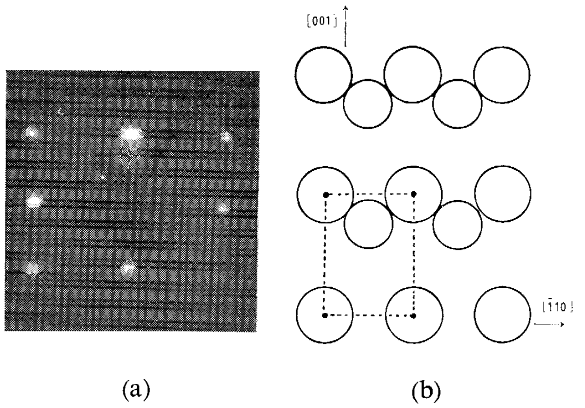
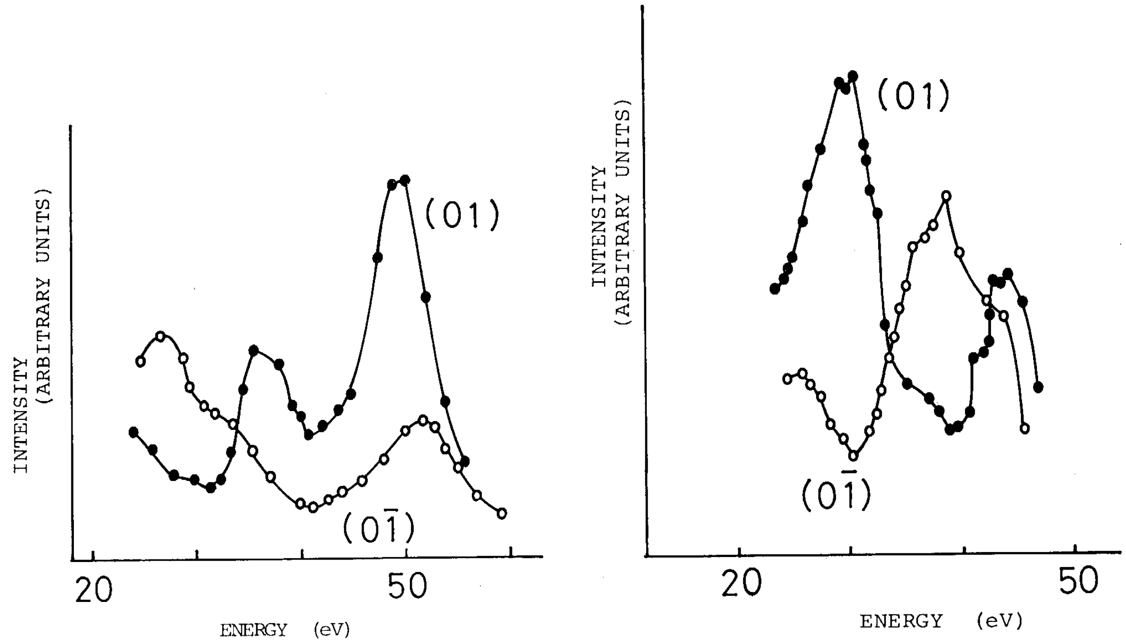


Fig. 2 A LEED pattern obtained from clean GaP (110) surface.
(a) Photographed at 32 eV.
(b) Atomic arrangement of a unit cell of the GaP (110) surface.



(a) Fig. 3 Intensity distribution curves. (b)

(a) for GaAs (110) (b) for GaP (110)

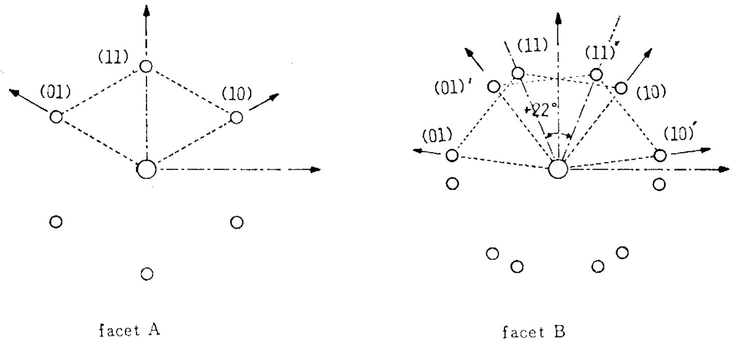
3. Epitaxial growth of Al on GaAs

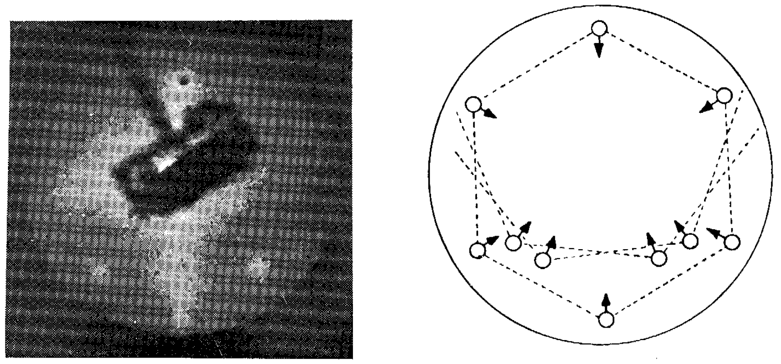
3.1 Films deposited at high substrate temperatures

LEED patterns obtained at early stages of the deposition at all substrate temperatures (R.T.~200°C) showed essentially the same behavior: GaAs spots gradually faded out in the increased background as the deposition to leave only a bright background at about 30 Å exposure.

Further exposure of Al over 50 Å caused a faint and long streak along the [001] axis of the substrate. The streak was visible only below 45 eV, whose non uniform intensity distribution suggested a facet characteristics. Slight addition of Al to this stage produced the diffuse spots which became sharper after further deposition. LEED pattern at this stage is shown in Fig. 4 with the illustration. In the illustration short arrows attached to each spots indicate the direction of the movement of spots when the incident beam energy is increased, however, all spots converge to two points A, B other than the original (00) spot. This suggests all spot are due to two facets A, B.

Identification of the facet was made as follows; (1) by tilting the specimen so as to face the facet surface normal to the incident beam and observe the symmetry of diffraction spots and their spacing, and (2) to determine the tilt angle of the facet, a series of diffraction pattern was recorded by varying the incident beam energy under normal incidence to the substrate, and the diffraction spots were plotted in the reciprocal lattice space.





(a) (b)

Fig. 4 A LEED pattern from the Al film deposited (thickness 80 Å) deposited at high substrate temperature.

(a) Photographed at 41 eV.

(b) A schematic diagram. Spots moved to the direction by increasing incident electron energy.

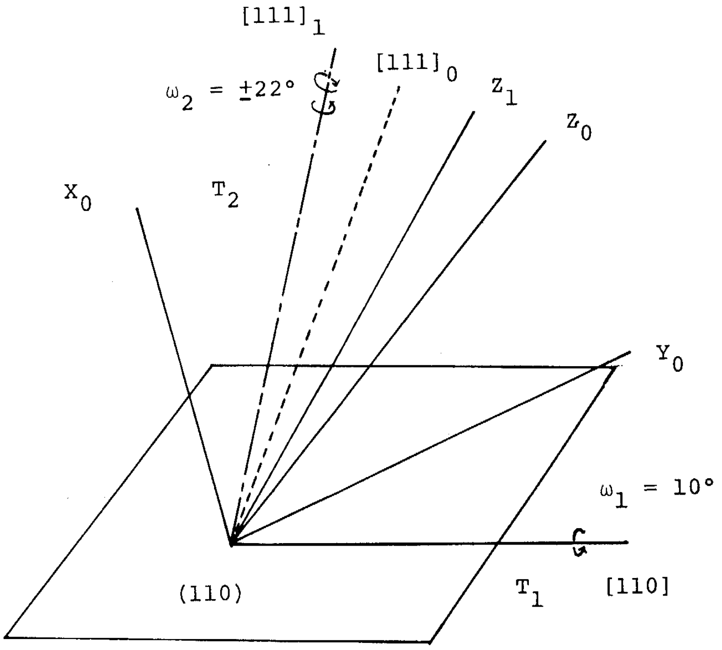


Fig. 5 A schematic diagram for transforming coordinates of substrate into those of crystallite.

The first procedure revealed that both facet A and B were Al (111) shown in Fig. 4. The (111) symmetry of the facet A can easily been seen in Fig. 4 where (11) of the two dimensional unit mesh of Al (111) lies along [001] direction of the substrate. The same (111) symmetry is found in the facet B as shown in Fig. 4, but there are two orientations different from the facet A; the (11)'s locate at positions rotated 22° from the [001] axis of the substrate.

The second procedure revealed the tilt angle of 18°±0.5° for the facet A, and the angle of 25°±0.5° for facet B, zone axis of both facet A and B being parallel to [$\bar{1}$10] of the substrate. The tilt angle 18°±0.5° for the facet A allows one to assign (112) for the basal plane parallel to the substrate surface since the angle between (112) and (111) is 19.5°. In the case of facet B, the basal plane parallel to the substrate may be indexed by transforming the [110] vector of the substrate into the coordinate fixed to the crystallite having the facet B.

Refering Fig. 5, this transformation can be performed as,

$$\begin{pmatrix} x \\ y \\ z \end{pmatrix} = T_2^{+} T_1 \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix}$$

$$= \begin{pmatrix} 1-\frac{2}{3}\kappa_2 & \frac{1}{3}\kappa_2+\frac{\sqrt{3}}{3}\sin\omega_2 & \frac{1}{3}\kappa_2-\frac{\sqrt{3}}{3}\sin\omega_2 \\ \frac{1}{3}\kappa_2-\frac{\sqrt{3}}{3}\sin\omega_2 & 1-\frac{2}{3}\kappa_2 & \frac{1}{3}\kappa_2+\frac{\sqrt{3}}{3}\sin\omega_2 \\ \frac{1}{3}\kappa_2+\frac{\sqrt{3}}{3}\sin\omega_2 & \frac{1}{3}\kappa_2-\frac{\sqrt{3}}{3}\sin\omega_2 & 1-\frac{2}{3}\kappa_2 \end{pmatrix}$$

$$\begin{pmatrix} 1-\frac{\kappa_1}{2} & -\frac{\kappa_1}{2} & -\frac{1}{\sqrt{2}}\sin\omega_1 \\ -\frac{\kappa_1}{2} & 1-\frac{\kappa_1}{2} & -\frac{1}{\sqrt{2}}\sin\omega_1 \\ \frac{1}{\sqrt{2}}\sin\omega_1 & \frac{1}{\sqrt{2}}\sin\omega_1 & 1-\kappa_1 \end{pmatrix} \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix}$$

$$= \begin{pmatrix} 4.001 \\ 2.96 \\ 1 \end{pmatrix} \simeq \begin{pmatrix} 4 \\ 3 \\ 1 \end{pmatrix}$$

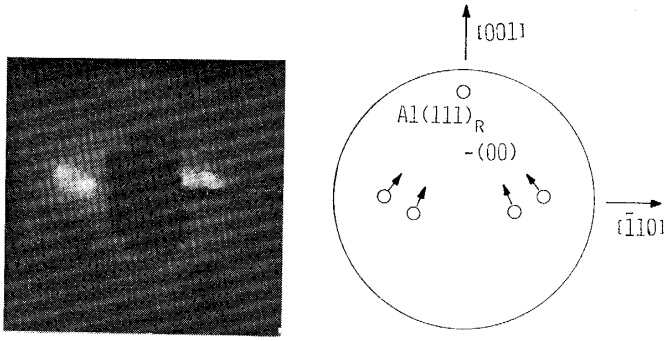
$$\begin{pmatrix} x \\ y \\ z \end{pmatrix} = T_2^{-} T_1 \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix} \simeq \begin{pmatrix} 3 \\ 4 \\ 1 \end{pmatrix}$$

Where, $k_1=1-\cos\omega_1$, $k_2=1-\cos\omega_2$, $\omega_1\simeq 10°$, $\omega_2=\pm 22°$.

Thus the basal plane may be indexed as (431) and (341). The facts that (00)'s of both facet A and B were observed only on the positive side of [001] axis of the substrate and that there were no exposed Al(110) lead to asymmetric faceting.

3.2 Films deposited at room temperature

In this case, a definite LEED pattern obtained from Al exposure of about 60 Å is shown in Fig. 6, below 60 Å only an intense background being observed. The LEED pattern shown in Fig. 6 clearly shows the Al(111) facet having the same orientation as the facet B described above, but the tilt angle was determined to be 35°±0.3°. In addition, the facet tilts towards positive [001] direction as in the high temperature case. The basal planes parallel to the substrate were tentatively indexed as (24 15 4) and (15 24 1) following above mentioned method. Annealing the films over 400°C for several second produced diffraction spots having GaAs(110) symmetry in a reduced background, but the nature of this surface was not studied further.



(a) (b)

Fig. 6 A LEED pattern from the Al film deposited (thickness 60 Å) deposited at room temperature.
(a) Photographed at 59 eV.
(b) A schematic diagram. Spots moved to the direction by increasing incident electron energy.

4. Epitaxial growth of Al on GaP

In low substrate temperature case, an increased background made the initial LEED pattern hardly visible until the Al exposure of about 50 Å, however, some new diffuse spots appeared after the deposition of above 50 Å. When the thickness was increased over 100 Å, a relatively difinite pattern was obtained. The LEED pattern clearly shows the existence of (111) facet having the same orientation as the facet B described above and the tilt angle takes the value of 25°±0.5°. The planes parallel to the substrate were tentatively indexed as (431) and (341) as the system Al–GaAs case. In high substrate temperature case, the LEED pattern shows essentially the same behavior except of {111} facets as compared with the low temperature case.

5. Conclusion

From the results of the present study, remarkable features in two systems are evident. (1) An asymmetry in the intensity of (01) and $(0\bar{1})$ spots was found in the LEED observation for GaAs(110) and GaP(110). (2) The asymmetric film structure should be emphasized in both systems Al–GaAs and Al–GaP. (3) At high temperatures (high *Ts*), the (112) base of (111) facet is only found in Al–GaAs. (4) At low *Ts*, (111) facet of Al–GaAs have the (25 14 1), (14 25 1) base, while those of Al–GaP have the (431), (341) base. An asymmetric film structure found out in the present study was also observed in the system Ag–GaAs(110), Au–GaAs(110) and Au–GaP(110); only $(35\bar{1})$ and $(53\bar{1})$ facets are formed but no (351) and (531) in the system Ag–GaAs and Au–GaAs at low *Ts*. Only (111) facet are formed but no $(11\bar{1})$ in Au–GaP at low *Ts*. This feature is thus thought to be common when films are grown on cleaved surfaces of zincblende type crystals. It is certain that the asymmetry reflects the asymmetric atomic arrangement in the substrate surface along the [001] axis. Moreover, the asymmetric film structure should be emphasized on the film deposited at low *Ts*. An asymmetric film structure found out in the present study was also observed in Ag–O systems8)9).

Acknowledgement

The author would like to thank professor T. Hanawa of Electron Beam Laboratory, Faculty of Engineering, Osaka University for useful discussions in the present study.

References

1) M. Kubo and S. Miyake: J. Phys. Soc. Japan **3**, 114 (1948)
2) N. Miyata and T. Hanawa: Japan J. Appl. Phys. Suppl. **2** Pt. 1, 593 (1974)
3) T. Hanawa and K. Takeda: Acta. Crystallogr. **A28** S145 (1972)
4) K. Takeda, T. Hanawa and T. Shimojo: Japan J. Appl. Phys. Suppl. **2** Pt. 1, 589 (1974)
5) N. Miyata and T. Hanawa: to be published
6) H. C. Gatos: "Progress in semiconductors", eds. Gibson, Kroger and Burgess (John Wiley, N. Y.) vol. 9 (1965)
7) A. U. MacRae and G. W. Gobeli: J. Appl. Phys. **35**, 1629 (1964)
8) N. A. Gjostein: Acta. Meta. **11**, 969 (1963)
9) A. J. W. Moore: Acta. Meta. **6**, 293 (1958)