Preferential Growth of Al (110) Films on GaAs (100) Surfaces under Rapid Vapor Deposition in Ordinary High Vacuum

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In order to confirm the effect of the rapid evaporation during film growth, 100 nm thick Al films were deposited from dual sources of W baskets onto (100) surfaces of GaAs substrates heated at a temperature of 473 to 723 K in an ordinary high vacuum (10^{-3} Pa). From a transmission electron microscopic examination of the films, it was found that the optimum substrate temperature was 673 K for the growth of a (110) epitaxial film; the obtained film was mostly composed of islands with the orientation relationship of $(110)[001]_{\text{Al}}// (100)[011]_{\text{GaAs}}$. During the early stage of deposition, however, numerous islands grew in various crystallographic orientations on the substrate heated at 673 K; the epitaxy was found in the films thicknesses greater than 80 nm.

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1. Introduction

Several investigators have reported the epitaxial growth of Al on a GaAs (100) surface by molecular beam evaporation (MBE) in an ultra-high vacuum. Ludeke *et al.*¹⁾ first found that an Al (110) film grew epitaxially with the orientation relationship of $(110)[001]_{\text{Al}}// (100)[011]_{\text{GaAs}}$ at a growth rate of 0.001 to 0.1 nm/s where the substrate was heated at a temperature of 323 to 773 K. According to Cho and Dernier,²⁾ an Al (100) film also developed on the substrate at room temperature with the relationship of $(100)[001]_{\text{Al}}// (100)[011]_{\text{GaAs}}$ at a growth rate of 0.06 nm/s. On the contrary, Massies *et al.*³⁾ pointed out that Al (110) or (100) growth was observed, depending on the specific conditions of growth temperature and growth rate, and that the Al [001] axis was parallel to either the GaAs [011] or $[0\bar{1}1]$ directions (double positioning). Consequently, practical epitaxy seems to be very complex for the film growth of Al on a GaAs substrate by MBE.

On the other hand, it is believed that an oriented film of Al is difficult to grow on a GaAs substrate when vapor deposition is carried out in an ordinary high vacuum. The present author, Sugawara,⁴⁾ found that an Al (100) film grew epitaxially on a cleaved NaCl crystal (623 K) in an ordinary high vacuum of 2×10^{-3} Pa at a high deposition rate of 3–6 nm/s using dual sources of W-baskets. It is the purpose of this study to examine the effect of rapid evaporation of Al from dual sources on crystallinity of the films which grow on a (100) surface of GaAs substrate heated in an ordinary high vacuum.

2. Experiments

A GaAs (100) wafer, 77 mm in diameter and 0.6 mm in thickness, was obtained from an undoped single crystal grown by the liquid encapsulated Czochralski (LEC) method and was cut along the $\langle 011 \rangle$ directions into small square specimens with an 11 mm edge length. After being cleaned in acetone for 600 s using an ultrasonic cleaning equipment, the

specimen was chemically polished in a solution composed of 97% $\text{H}_2\text{SO}_4 + 30\% \text{H}_2\text{O}_2 + \text{H}_2\text{O}$ (4 : 1 : 1 volume ratio, room temperature) for 300 s, followed by etching with a solution of 36% $\text{HCl} + \text{H}_2\text{O}$ (1 : 10 volume ratio, room temperature) for 600 s. It was then washed in distilled water and mounted as a substrate in a sample holder in the heating stage within a bell jar of a vacuum system.

On the other hand, two small pieces of Al were cut from an ingot (99.999% in purity) as a material for evaporation. They were electropolished at an anodic voltage of 1.7 V in a solution of 60% $\text{HClO}_4 + \text{CH}_3\text{OH}$ (1 : 4 volume ratio, room temperature) and each piece, of the same weight, was put separately into the two neighboring baskets⁴⁾ which were made of 0.7 mm diameter W-wire. The dual baskets were separated by 2 cm from one another and they were placed under the substrate at a distance of about 10 cm. Subsequently, the substrate was heated to a temperature of 473 to 723 K in an ordinary high vacuum of 2×10^{-3} Pa and the simultaneous evaporation from the dual baskets was carried out to obtain 100 nm thick Al films. The deposition rate was 3–6 nm/s. Preliminary experiments showed that the time required for increasing the heating current through the baskets from 0 to 28 A was desired to be less than 5 s in order to produce the film with a preferred orientation. The crystallinity of the Al film was investigated by transmission electron microscopy (TEM). For the sake of inspection, the film was protected by a C coating in a vacuum and then separated from the substrate by preferential attack of the GaAs with a solution of 25% $\text{NH}_4\text{OH} + 30\% \text{H}_2\text{O}_2 + \text{H}_2\text{O}$ (1 : 1 : 5 volume ratio, room temperature). After being washed in a solution of 25% $\text{NH}_4\text{OH} + 30\% \text{H}_2\text{O}_2 + \text{H}_2\text{O}$ (1 : 1 : 20 volume ratio, room temperature) and in acetone, the fragments of the film were fixed on Cu grids and inspected using a transmission electron microscope (JEOL, JEM-2010), which was operated at an accelerating voltage of 200 kV; bright field images were photographed at a magnification of 2×10^4 and selected area diffraction (SAD) patterns were obtained from a $\phi 1.3 \mu\text{m}$ area of the Al film using an SAD aperture. After a survey of the

most appropriate growth temperature for the epitaxial growth, the thickness of the deposited Al films was changed from 40 to 250 nm at a substrate temperature of 673 K in order to examine the growth mode of the films. Inspection of their crystallinity was also carried out by TEM.

3. Results and Discussion

3.1 Effect of the substrate temperature

Figure 1 shows electron micrographs and SAD patterns ob-

Fig. 1 Transmission electron micrographs and SAD patterns which were obtained from the 100 nm thick Al films. The deposition was carried out from the dual sources of W-baskets onto the GaAs (100) substrates heated at 473 K ((a) and (b)), 573 K ((c) and (d)), 673 K ((e) and (f)) and 723 K ((g) and (h)) in an ordinary high vacuum.

tained from the Al films which were prepared by rapid evaporation from the dual sources onto the GaAs (100) substrates: the substrates were heated at temperatures of 473, 573, 673 and 723 K, respectively. The 100 nm thick Al films are composed of islands, which grow large with an increase in the substrate temperature and reach a maximum size of about $0.5\text{ }\mu\text{m}$ at 673 K (Fig. 1(e)). On the other hand, the preferred (110) orientation of Al is developed over a wide area at a growth temperature of 673 K (Fig. 1(f)), although no epitaxy is observed at the other growth temperatures examined. It should be noted that double positioning is quite absent in this film. Ludeke *et al.*¹⁾ suggested that such an absence might originate from the preferred occurrence of the tetrahedral Al-As bond at the interface between the Al deposit and GaAs substrate. As mentioned in the Introduction, the orientation relationship is $(110)[001]_{\text{Al}}// (100)[011]_{\text{GaAs}}$. In this relationship, the lattice misfit δ between $[001]_{\text{Al}}$ and $(1/2)[011]_{\text{GaAs}}$ is calculated to be $+2.0\%$ at 673 K.

For comparison, a single source W-basket was employed to evaporate Al to a thickness of 100 nm on the GaAs (100) substrate heated at 673 K. Figure 2(a) shows an electron micrograph of the deposited film. The size of the islands is about $0.4\text{ }\mu\text{m}$; it is rather small in comparison with the size of the islands which are observed in the film prepared under the similar experimental conditions, except for the use of dual sources (refer to Fig. 1(e)). Furthermore, the SAD pattern (Fig. 2(b)) clarifies the existence of islands with some different orientations, in particular, the preferred occurrence of the double positioning of the (110) islands with the orientation relationship of $(110)\langle 001 \rangle_{\text{Al}}// (100)\langle 011 \rangle_{\text{GaAs}}$.³⁾ For an understanding of the double positioning, Fig. 2(b) shows a diagram which compares the superimposition of two $\{110\}$ diffraction patterns rotated $\pi/2$ rad to one another. Accordingly, it can be con-

cluded that the rapid evaporation from dual sources is quite effective for growing an Al (110) epitaxial film on a GaAs (100) substrate heated in an ordinary high vacuum (refer to Figs. 1(e)–(f)).

3.2 Effect of the thickness of deposited Al films

Figure 3 shows the changes in the morphology and the crystallinity of the Al films with an increase in the thickness from 40 to 250 nm. At an early stage of vacuum deposition, many islands nucleate in various crystallographic orientations (Figs. 3(a)–(d)). The increase in the film thickness to 80 nm results in an enlargement of the islands of preferred (110) orientation along with the coalescence of neighboring islands (Figs. 3(e)–(f)), until the (110) islands impinge on each other and grow into large subgrains (Figs. 3(g)–(h)). Such a growth mode is quite similar to the one which was observed in the epitaxial growth of an Al (100) film⁴⁾ on the cleaved (100) surface of a NaCl crystal under high rate deposition from dual sources in an ordinary high vacuum, except for the formation of many sub-boundaries and some grain boundaries in the present study.

However, our attempt could not clarify the mechanism of the epitaxial growth of an Al film by rapid deposition from dual sources. As suggested in a previous paper,⁴⁾ it is assumed that the growth of nuclei may be suppressed by the segregation of oxygen species attached on the surface of nuclei when the vacuum evaporation from a single source is carried out in an ordinary high vacuum. If the segregation of oxygen species is neglected even in an ordinary high vacuum, the faster growth of nuclei and their coalescence into islands with the preferential orientation will occur by adopting a rapid evaporation from dual sources to the properly heated substrate, thus resulting in the growth of an epitaxial Al film.

Fig. 2 Transmission electron micrograph and SAD pattern which were obtained from the 100 nm thick Al film. The deposition was carried out from the single source of a W-basket onto the GaAs (100) substrates heated at 673 K in an ordinary high vacuum.

Fig. 3 Transmission electron micrographs and SAD patterns which were obtained from the Al films with thicknesses of 40 nm ((a) and (b)), 60 nm ((c) and (d)), 80 nm ((e) and (f)) and 250 nm ((g) and (h)). The deposition was carried out from the dual sources of W-baskets onto the GaAs (100) substrates heated at 673 K in an ordinary high vacuum.

4. Conclusion

The 100 nm thick Al films were rapidly deposited from

dual sources of W-baskets onto the GaAs (100) substrates heated at temperatures from 473 to 723 K in an ordinary high vacuum of 2×10^{-3} Pa at the deposition rate of 3–6 nm/s. After being reinforced with a carbon coating in a

vacuum and separated from the substrates by chemical etching, the films were examined by transmission electron microscopy. As a result, it was found that the film deposited at a substrate temperature of 673 K was composed of islands the size of about 0.5 μm with the orientation relationship of $(110)[001]_{\text{Al}}// (100)[011]_{\text{GaAs}}$; no epitaxial growth occurred at the other substrate temperatures.

When the thickness of the Al films deposited from dual sources was increased from 40 to 250 nm at a growth temperature of 673 K, numerous islands first nucleated in various crystallographic orientations and then grew large in the preferred (110) orientation along with the coalescence of neighboring islands at film thicknesses greater than 80 nm until the (110) islands impinged on each other and grew into large subgrains. Based on these facts, it is assumed that rapid evaporation is essential for the epitaxial growth of Al films in order to prevent the segregation of oxygen species on the surface of

the islands.

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