Epitaxial Growth of Stoichiometry-Controlled CoAl Films on AlAs(001)

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CoAl films were grown on AlAs(001) by molecular beam epitaxy. By strict control of stoichiomety of Co and Al, a strong (2x2) reconstruction was obtained on MBE-grown CoAl(001) surfaces. These surfaces are found to be Al-terminated surfaces. AlAs were grown on both Al-terminated and Co-terminated CoAl(001) surfaces. AlAs(001) single phase was found to grow on Co-terminated CoAl(001) surfaces, while a mixed-phase of AlAs(001) and AlAs(111) was found to grow on Al-terminated surfaces.

1. Introduction

Epitaxial metal/semiconductor heterostructures are the basic components of semiconductor/metal /semiconductor heterostructures which have high potentiality for new device applications and novel physics. Intermetallic compounds such as NiAl, NiGa, CoAl and CoGa have been reported to grow on AlAs,¹⁾⁻⁵⁾ because these intermetallic compounds have a cubic CsCl structure and their lattice constants are almost equal to half the lattice constants of GaAs and AlAs. Among these intermetallic compounds, CoAl (lattice constant is 2.861Å) has the smallest lattice mismatch to AlAs (1.1%) and has the highest melting point (1648°C), hence, is attractive as constituent material in epitaxial metal/semiconductor heterostructures.4)

We study molecular beam epitaxial growth of CoAl films on AlAs(001), where the stoichiometry of Co and Al is strictly controlled. We pay special attention to the characteristics of intrinsic CoAl surfaces: besides surface reconstructions, we examine the kinds of the terminated surfaces of CoAl films (Co or Al-terminated). Understandings of the surface characteristics of metal films are important when we grow semiconductors on epitaxial metal films. We also study the growth of AlAs on CoAl films taking account of the surface characteristics of CoAl films.

2. MBE Growth of CoAl on AlAs(001)

CoAl films were grown on 200Å AlAs(001) layers which were grown by conventional molecular beam epitaxy (MBE). The growth temperature of CoAl was Ts=350°C and the backgrownd chamber pressure was below $3x10^{-10}$ Torr which is low enough for the

high-purity metal atom deposition. Molecular beam intensity of Co and Al was calibrated with an accuracy of 1% by element-luminescence analysis using inductively coupled plasma. The composition of Co and Al was therefore controlled within an error of 1-2%. In the growth of CoAl films, a single monolayer(ML) of Co atoms were first deposited on AlAs(001) surface, then Co and Al atoms were co-deposited onto the surface. The growth rate was 1ML/min.

Figure 1(a)-(c) are the reflection high-energy electron diffraction (RHEED) patterns ([110] azimuth) observed in the growth of CoAl films, where the stoichiometry of Co and Al was strictly controlled within an error of 1-2%. Figure 1(a) is the pattern of As-stabilized AlAs(001) surface with a (x2)reconstruction. When a single monolayer of Co atoms were deposited on As-stabilized AlAs(001) surface, the RHEED pattern was observed to be changed to the pattern of Fig.1(b), whose streak intervals correspond to the lattice constant of CoAl. After the growth of 2-3ML CoAl, a strong surface reconstruction of (x2) pattern was observed to appear. This surface reconstruction was maintained throghout the growth. Figure 1(c) shows the (x2) reconstruction on 200Å CoAl film. The (x2) reconstruction was also observed at the $[\overline{110}]$ azimuth. Figure 2 is the rocking curve of X-ray diffraction for the 200Å CoAl film on AlAs(001). CoAl(001) and (002) peaks were observed. Thus, RHEED observation and X-ray diffraction measurements show that CoAl(001) single crystal films were grown on AlAs(001) surfaces with the epitaxial relationship of CoAl(001)[110]//AlAs(001)[110].

We also grew CoAl films whose composition of Co (or Al) is deviated from 50% by several percents. The surface reconstruction of (2x2) was not observed when the composition of Co is 54% (Co-rich) or the composition of Al is 53% (Al-rich), although X-ray



(a) As-stabilized AlAs(001)



(b) Co(1ML)



(c) 200Å CoAl

Fig.1 RHEED patterns in the growth of CoAl films ([110] azimuth).

diffraction measurements showed the formation of CoAl(001) single crystalline. Thus, the (2x2) surface reconstruction was found to appear when the stoichiometry of Co and Al is strictly controlled, and this surface reconstruction is considered to be the intrinsic one of the CoAl(001) surface.

For the MBE growth of intermetallic compounds, kinds of the grown surface terminations have never been discussed. We examined whether the MBE-grown CoAl surfaces have the Co-terminated or Al-terminated surface. We deposited 1ML of Co onto the MBE-grown CoAl(001) surfaces, and observed changes in the RHEED pattern. Figure 3(a)-(c) shows the results. Figure 3(a) is the RHEED pattern ([110] azimuth) of a MBE-grown CoAl(001) surface which shows a strong (x2) reconstruction. When 0.5ML of Co was deposited on this surface, the (x2) reconstruction was observed to disappear (Fig.3(b)). However, a strong (x2) reconstruction was observed to recover by subsequent deposition of Co (total 1.0ML Co)(Fig.3(c)). On the other hand, when we deposited 1ML of Al instead of Co onto the MBE-grown CoAl(001) surfaces, such changes in RHEED pattern were not observed: after the (x2) reconstruction desapeared (0.5ML Al), (x1) streaks of CoAl was observed to be gradually decreased. Moreover, semi insitu Auger electron measurements using LVV peaks of Co(55eV) and Al(65eV) showed the evidence that MBE-grown CoAl(001) surfaces are covered by Al atoms. Hence, we conclude that MBE-grown CoAl(001) surfaces have the Al-terminated surface.



Fig.2 X-ray diffraction spectrum (200Å CoAl/200Å AlAs(001)/GaAs(001))



(a) MBE-grown CoAl(001)



(b) 0.5ML Co deposition



(c) 1.0ML Co deposition

Fig.3 Changes in RHEED patterns ([110] azimuth) in the process of 1ML Co deposition on MBE-grown CoAl(001).

3. MEE Growth of AlAs on CoAl

In order to fabricate AlAs/CoAl/AlAs heterostructures, we grew AlAs on CoAl(001) films by migration-enhanced epitaxy (MEE). We prepared two kinds of CoAl(001) surfaces, *i.e.* Al-terminated and Coterminated surfaces, and studied the growth of AlAs on each of the surfaces. We used 56Å MBE-grown CoAl(001) films as Al-terminated surfaces, while we prepared Co-terminated surfaces by depositing 1ML of Co atoms onto the 56Å MBE-grown CoAl(001) films. Both kinds of CoAl surfaces were observed to show strong (x2) surface reconstructions. The growth temperature of AlAs was Ts=450°C for both cases.



(a) 200Å AlAs/56Å Al-stabilized CoAl(001)



(b) 200Å AlAs/56Å Co-stabilized CoAl(001)

Fig.4 RHEED patterns of AlAs on CoAl(001) films.

Figure 4(a) and (b) are the RHEED patterns ([110] azimuth) of 200Å AlAs films grown on (a) Alterminated and (b) Co-terminated CoAs(001) surfaces. In Fig.4(a), in addition to AlAs(001) spotty pattern, AlAs(111) spotty pattern is also observed, indicating the growth of a mixed-phase of AlAs(001) and AlAs(111). On the other hand, in Fig.4(b), only AlAs(001) spotty pattern is observed, indicating the growth of a single phase of AlAs(001). Figure 5(a) and (b) are the rocking curves of X-ray diffraction for the structure of GaAs(1000A)/AlAs(200A)/CoAl(56A) /A1As(200Å)/GaAs; (a) and (b) corespond to the growth on Al-terminated and Co-terminated CoAl(001) surfaces, respectively. In Fig.5(a), in addition to AlAs(002) and (004) peaks, AlAs(111) peak is observed . In Fig.5(b), on the other hand, only AlAs(002) and (004) peaks are observed, and AlAs(111) peak is not observed. These results of X-ray diffraction are consistent with the RHEED observation. Thus, a mixed-phase of AlAs(001) and AlAs(111) are shown to grow on Al-stabilized CoAl(001) surfaces, while a single phase of AlAs(001) are shown to grow on Co-terminated CoAl(001) surfaces. We suppose that the surface Co layer weakens the bond strength between CoAl and AlAs, and reduces the growth of AlAs(111) phase which might result from strong combination of CoAl and AlAs.

4. Conclusions

By strict control of stoichiometry of Co and Al, MBE-grown CoAl(001) surface with a strong (2x2) reconstruction can be obtained. This surface is found to be the Al-terminated CoAl(001) surface. The Coterminated CoAl(001) surface which can be produced by the deposition of 1ML Co is also found to show a strong (2x2) reconstruction. The Co-terminated CoAl(001) surface is suitable for the AlAs overgrowth, because only AlAs(001) phase grows on this surface while both AlAs(001) and AlAs(111) phases grow on Al-terminated CoAl(001) surface.





(b) AlAs & GaAs on Co-terminated CoAl(001) film

Fig.5 X-ray diffraction spectra (1000ÅGaAs/200ÅAlAs/56ÅCoAl(001)/200Å/AlAs /GaAs(001))

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