PERIODICA POLYTECHNICA SER. CHEM. ENG. VOL. 50, NO. 1, PP. 13–20 (2006)

GROWTH OF $InAs_xSb_{1-x}$ LAYERS ON GaAs SUBSTRATES BY HOT WALL EPITAXY

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Received: Sept. 14, 2004

Abstract

We have studied the structural and electrical characteristics of InAsSb ternary layers grown on GaAs (001) substrates by hot wall epitaxy (HWE) at arsenic (As) reservoir temperature in the range from 220 to 290 °C. The growth rate of the epilayer is found to be decreased with increasing As temperature. This is attributed to the abundance of group V molecules to the growth surface, which suppresses the mass transport of Indium (In) atoms. A dramatic change in the surface morphologies of the samples has been observed by scanning electron microscopy. X-ray diffraction studies indicate that the arsenic composition of the layer can be controlled by manipulating As temperature. Hall effect results of the samples show that the electron mobility of the layer decreases with increasing As temperature.

Keywords: hot wall epitaxy, growth from vapor, semiconducting III-V materials, surface morphology, X-ray diffraction.

1. Introduction

High performance III-V ternary semiconductor-based devices have been received much attention in the past few years due to their potential applications to infrared detectors, gas sensors and lasers operating in mid-infrared (2-5 μm) and long wavelength (8-12 μm) regions [1]-[4]. Epitaxial growth of $InAs_xSb_{1-x}$ ternary layers on III-V substrates is practical interest for such applications because of their superior performance over the dominant HgCdTe based devices in which non-uniformity and instability factors deteriorate the performance of the devices [5]. Besides, the cut-off wavelength of $InAs_xSb_{1-x}$ layers can be tuned in a wide range upon controlling the alloy composition. Due to the reason, many growth techniques have been used to grow InAsSb ternary layers on III-V substrates. Recently, melt epitaxy (ME) has been employed for the growth of $InAs_xSb_{1-x}$ layers on InAs substrate [6, 7]. In this technique, the control of thickness of the grown layer is difficult owing to rapid growth rate of the layer. Likewise, $InAs_xSb_{1-x}$ layers grown by vapor phase epitaxy (VPE) have inferior quality due to low vapor pressure of In precursor compared to As and Sb precursors.

To date, non-equilibrium and expensive techniques such as molecular beam epitaxy (MBE) or metal organic chemical vapor deposition (MOCVD) techniques have been used to grow ternary alloy semiconductors [8]-[10]. On the other hand, hot wall epitaxy (HWE) is a simple technique and the epitaxial growth can be proceeded near thermodynamic equilibrium conditions. The technique has been successfully used to grow high quality epilayers of II-VI compounds [11, 12] and GaN [13]. However, there is no detailed report on the growth of InAsSb ternary layers using HWE. In this work, we report the growth of InAsSb layers on GaAs (001) substrates and propose a technique to control the arsenic composition. Structural and electrical characteristics of the layers have been analysed using scanning electron microscopy (SEM), X-ray diffraction (XRD) and Hall effect techniques.

2. Experimental Details

In the present investigations, InAsSb layers were grown on semi-insulating GaAs (001) substrates by hot wall epitaxy with triple cell configurations. Before the layer growth, the substrates were etched in H_2SO_4:H_2O_2:H_2O (3:1:1 vol. %) for 90 sec followed by ultrasonically cleaning with acetone and ethanol. After the etching, the substrates were rinsed with deionized H_2O and dried with N_2 gas flow subsequently loaded into the growth chamber. Indium (In), antimony (Sb) and arsenic (As) solids were used as source materials. The substrate, In and Sb temperatures were kept at 500, 780 and 470 °C, respectively. A series of InAsSb samples were grown at various As temperatures in the range from 220 to 290 °C at an interval of 10 °C. The substrate and source temperatures were controlled by an independent thermocouples. Prior to the layer growth, substrate and source heat treatments were performed for 60 minutes. For all the samples, the layer growth time employed was 2 hours.

Surface morphologies of the grown layers were analysed by SEM (JSM-6360LA). XRD (RINT2000) measurements of (400) plane reflections of CuKα radiation (λ = 1.54 Å) were carried out to analyse the structural characteristics and to estimate the arsenic composition of the layer. Electron mobility of the samples was estimated by Hall effect (HL5500PC) measurements using Van der Pauw technique. All the measurements were carried out at 300 K.

3. Results and Discussion

Fig. 1 shows the V/III ratio of vapor pressures of the solid sources as a function of As temperature. It is observed that the ratio is increased monotonically with increasing As temperature. Since the In and Sb temperatures remain unchanged, the increase of V/III ratio is attributed to the increase of As vapor pressure and it is more pronounced at high As temperature. *Fig. 2* shows the V/III ratio dependence on the growth rate of the $InAs_xSb_{1-x}$ layers. The growth rate of the layer is estimated

from the thickness of the layer. A rapid decrease in the growth rate of the layers is observed with increasing the V/III ratio until the value of 127, which corresponds to As temperature of 260 °C. It is considered that when increasing As temperature up to 260 °C, As concentration on the growth surface is increased due to the increase of As vapor pressure. As a consequence, an abundance of As molecules associated with Sb molecules to the surface of GaAs suppresses the mass transport of In adatoms to the growth layers, and therefore, the growth rate of the layer is decreased. In contrast to MOCVD growth technique [14], our results clearly suggest that the growth rate of InAsSb layers depends on the V/III ratio up to As temperature of 260 °C. At the higher values of V/III ratio, there is no further decrease in the growth rate of the layer, which is attributed to intensive suppression of the mass transport of In atoms to the growth surface.

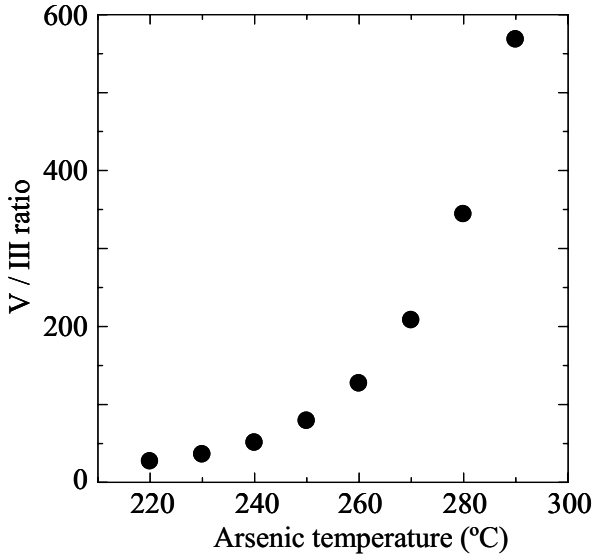

Fig. 1. The V/III vapor pressure ratio of the solid sources as a function of As temperature.

Surface morphologies of the layers grown at As temperature of 220, 240, 260 and 280 °C are shown in *Figs. 3(a)-(d)*. SEM images reveal that plateau-like 2D crystals nucleate at the As temperature of 220 °C (*Fig. 3(a)*). We consider the growth of the layer follows volmer-weber growth due to the large lattice mismatch between the growth layer and the substrate. The lattice mismatch of InAsSb layer on GaAs is between 14.6 and 7.2 % depending upon the arsenic composition. When increasing As temperature, the incorporation of As atom could cause relaxation of compressive strain in the layer, and the surface energy of the layer decreases due to stronger In-As atom bond energy than In-Sb atoms. As a result, quasi 2D crystals have been observed at 240 °C (*Fig. 3(b)*). As can be seen in *Fig. 3(c)*, high

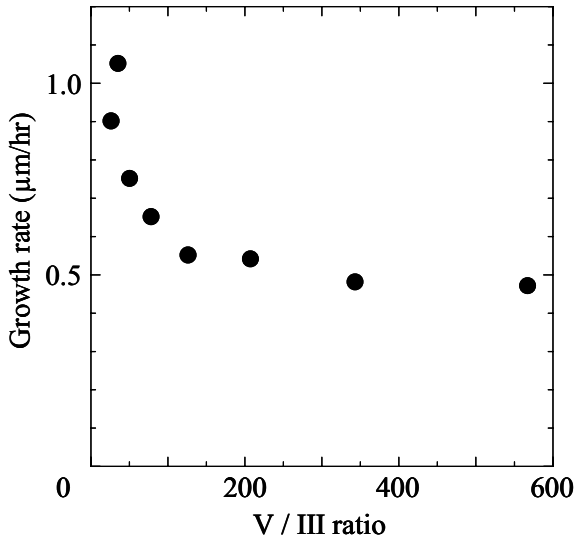

Fig. 2. The V/III vapor pressure ratio dependence of the growth rate of the $InAs_xSb_{1-x}$ layer.

density of coalesced 3D islands can be observed at 260 °C. The crystals initially nucleate plateau-like crystals followed by 3D islands growth without the formation of wetting layer. In this case, the surface energy between the layer and substrate is further decreased. At 280 °C, the crystals are elongated in a preferred growth direction, which implies that a different incorporation behaviour of the atoms to the growth surface (*Fig. 3(d)*). Furthermore, it is anticipated that As atoms are increasingly accumulated on the growth surface of the layers at 280 °C.

X-ray diffraction measurements show that the grown layers were single crystal. The XRD intensity diffraction peaks of the samples grown at various As temperatures are shown in *Fig. 4*. The (400) plane of the bulk InSb and InAs crystals is indicated for reference at the 2θ value of 56.78 and 61.14 degree, respectively. The diffraction peaks of the InAsSb layers are shown between the peaks of the bulk crystals. It is observed that the peak positions shift towards the higher 2θ value by increasing As temperature. It is ascribed to the increase of As composition (x) in the $InAs_xSb_{1-x}$ layer.

Fig. 5 shows the estimated As composition (x) and X-ray intensity of the layers as a function of As temperature. The As composition (x) of the layer is calculated using Vegard law.

$$x = (a_{\mathrm{InSb}} - a_G)/(a_{\mathrm{InSb}} - a_{\mathrm{InAs}}), \tag{1}$$

where, a_{InSb} and a_{InAs} are the bulk lattice constants of InSb and InAs crystals, respectively. a_G is the lattice constant of the growth layer determined from the

XRD measurements. From the *Eq.*1, the estimated x values of the layers are found to be increased from $x = 0.02$ to 0.90 by increasing As temperature from 220 to 290 °C. Our results suggest that the arsenic composition of the $InAs_xSb_{1-x}$ layer can be controlled by adjusting As temperature, which is a key technique to control the wavelength of optical devices. In *Fig. 5*, it is noted that the x values are not linear with As temperatures. SEKI et al. [14, 15] analysed the solid compositions of III-V alloy semiconductors using thermodynamic equilibrium model. It has been reported that the solid composition ratio of the III-V-V alloy depends on the V/III ratio and Gibbs free energy. Besides, when V/III ratio is nearly equal to one, the composition of the growth layer varies linearly with the ratio of As to Sb. On the other hand, the V/III ratio is greater than one; the composition depends on Gibbs free energy and varies with a non-linear function. In our experiment, the V/III ratios at all temperatures are greater than one. The results suggest that the relation between the ratio of As to Sb and the composition of the grown layers is a non-linear function and is governed by Gibbs free energy.

As can be seen in *Fig. 5*, the XRD peak intensity is decreased and reaches a low value at 260 °C, and then increases with increasing As temperature. These results clearly indicate that the structural changes in the surface morphology of the layers influence the XRD intensity. In the range from 220 °C to 260 °C, the XRD peak intensity decreases due to alloy disordering of the growth layers. At 280 °C, however, the reason for increasing the XRD is not yet clear. Further, we note that the XRD intensity is stronger at 220 °C than at 280 °C. This phenomenon suggests that Sb-rich layer has better quality than As-rich layer. Further investigations are necessary to fully understand the alloy ordering effect on the grown layers.

The room temperature Hall effect measurements of the InAsSb layer show that electrons are majority carriers for transport mechanisms of all the samples. *Fig. 6* shows the electron mobility of the samples as a function of As temperature. It is observed that the electron mobility of the samples decreases with increasing As temperature till 270 °C and then increases. The decrease in the electron mobility is due to the increase of surface roughness in addition to the dominant optical phonon scattering at room temperature [16, 17]. We quantitatively measured the average surface roughness of the layer and it was found to be 9.5 nm at 220 °C, whereas it reached a maximum of about 55.0 nm at 260 °C.

4. Conclusion

$InAs_xSb_{1-x}$ ternary layers were grown on GaAs (001) substrate by HWE technique at various As reservoir temperatures in the range from 220 to 290 °C. The decrease in the growth rate of the layer is attributed to suppression of mass transport of In atoms to the growth surface by group V molecules. SEM images reveal that the nucleation of coalesced 3D island at the As temperature of 260°C is considered to be decrease in the surface energy between the layer and substrate. XRD results show that As composition of the layer is increased from 0.02 to 0.90 when increasing

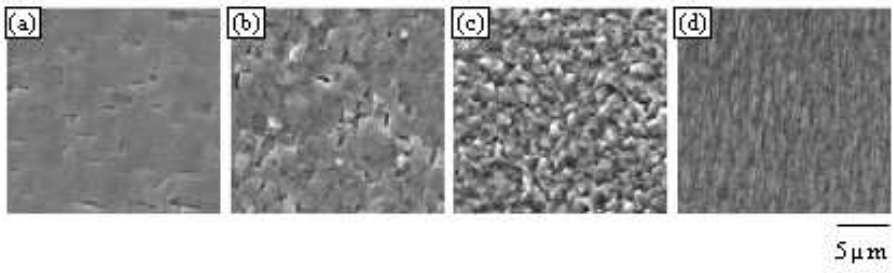

Fig. 3. SEM images of the samples grown at As temperatures of (a) 220, (b) 240, (c) 260 and (d) 280 °C.

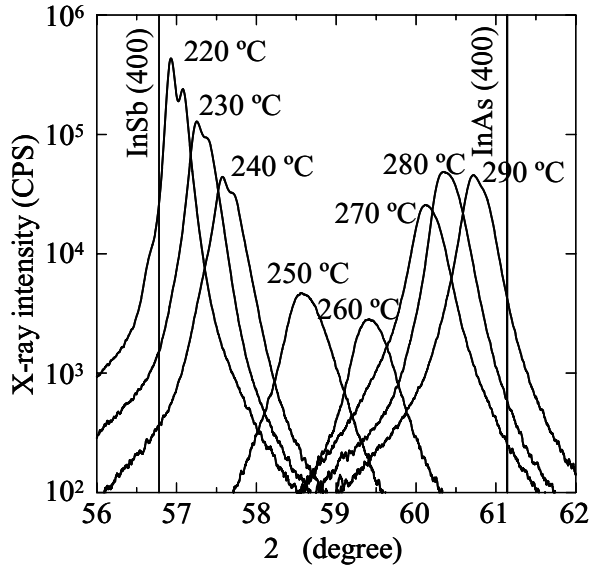

Fig. 4. Arsenic temperature dependence of X-ray diffraction peaks of the samples.

the As temperature from 220 to 290°C. This result suggests that As composition of the layer can be controlled in a wide range for practical applications by optimizing As temperature. The decrease in the electron mobility of the layers is caused by surface roughness of the layer.

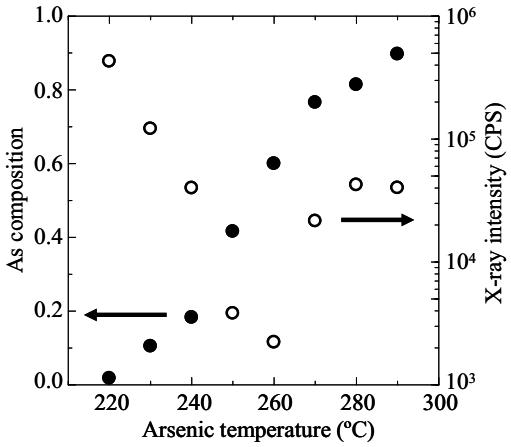

Fig. 5. Variation of As composition (x) and X-ray intensity of $InAs_xSb_{1-x}$ layer as a function of As temperature.

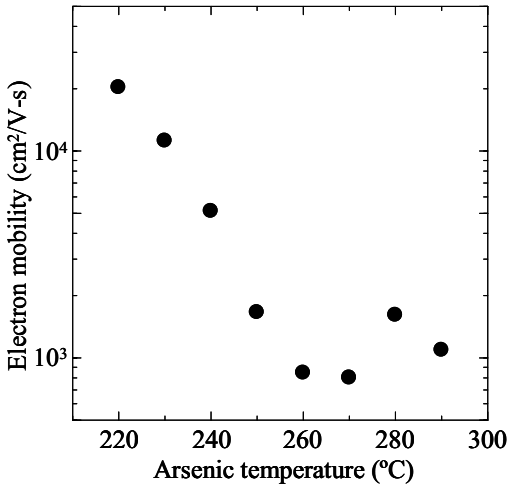

Fig. 6. The electron mobility of the samples as a function of the As temperature.

Acknowledgement

The authors would like to thank Prof. G. P. Srivastava (University of Exeter, U.K.) for useful discussions. We also thank Mr.Y.Momose for his help. We gratefully acknowledge Grand-in-Aid of Scientific Research from the Japanese Ministry of Education, Science and Culture (Kiban (B) No.14350164) for financial support to carry out this work.

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