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LOW-TEMPERATURE GROWTH CONDITIONS AND PROPERTIES OF $\text{AlGa}(\text{As})\text{Sb}$ ON GaSb SUBSTRATE BY LPE

S. Fujita, N. Hamaguchi, Y. Takeda and A. Sasaki

Department of Electrical Engineering, Kyoto University, Kyoto 606, Japan

Résumé. - La croissance par épitaxie en phase liquide des alliages semiconducteurs AlGaSb et AlGaAsSb sur le substrat GaSb a été étudiée en fonction de la température de croissance (T_G). A une température de croissance inférieure à 450°C , la morphologie et la vitesse de croissance des couches obtenues par épitaxie sont fortement influencées par le processus de préparation du substrat. Les couches ternaires obtenues par épitaxie $\text{AlGa}_{1-x}\text{Sb}$ avec $x=0,84$ sont obtenues avec succès sans instabilité d'interface entre les couches et le substrat en diminuant la température de croissance à 400°C . A une température de croissance de 540°C , le plus grand taux de composition en Al pour une couche ternaire sans instabilité d'interface est $\sim 0,64$ alors que, pour une couche quaternaire, il est $\sim 0,78$. Les couches quaternaires $\text{Al}_{0,78}\text{Ga}_{0,22}\text{As}_y\text{Sb}_{1-y}$ à interface droit peuvent être obtenues de manière sûre pour une température de croissance de 540°C . La concentration en porteurs est réduite d'une manière significative en abaissant la température de croissance : $p \approx 10^{17}\text{cm}^{-3}$ à $T_G=540^\circ\text{C}$ alors que $p \approx 8 \times 10^{15}\text{cm}^{-3}$ à $T_G=400^\circ\text{C}$. La photoluminescence des couches ternaires obtenues par épitaxie met en évidence quatre bandes d'émission distinctes à $4,2\text{K}$ probablement dues à la recombinaison radiative d'un exciton libre, d'un exciton lié à l'accepteur neutre et d'une paire D-A liée à 2 niveaux accepteurs différents.

Abstract. - Liquid-phase epitaxial growth of AlGaSb and AlGaAsSb alloy semiconductors on GaSb substrates has been investigated as a function of growth temperature (T_G). At a growth temperature lower than 450°C , the morphology and the growth rate of the epilayer are greatly affected by the substrate preparation processes. $\text{Al}_x\text{Ga}_{1-x}\text{Sb}$ ternary epilayer with $x=0.84$ is successfully grown without interface instability between the epilayer and the substrate by lowering the growth temperature to 400°C . At a T_G of 540°C , the largest Al-composition rate for a ternary layer without the instability is ~ 0.64 , whereas it for a quaternary layer extends to ~ 0.78 . $\text{Al}_{0.78}\text{Ga}_{0.22}\text{As}_y\text{Sb}_{1-y}$ layer with straight interface can be reliably grown at $T_G=540^\circ\text{C}$. Carrier concentration is significantly reduced by decreasing T_G : $p \approx 10^{17}\text{cm}^{-3}$ at $T_G=540^\circ\text{C}$, whereas $p \approx 8 \times 10^{15}\text{cm}^{-3}$ at $T_G=400^\circ\text{C}$. Photoluminescence of the ternary layers exhibits four distinct emission bands at 4.2K which are presumably arisen from radiative recombination of free exciton, exciton bound to neutral acceptor and D-A pair related to two different acceptor levels.

1. Introduction. - Much attention has been given to $\text{AlGa}(\text{As})\text{Sb}$ ternary or quaternary alloy semiconductor as an attractive candidate material for optoelectronic device application in a $1\text{-}\mu\text{m}$ wavelength range because of its wide variation of bandgap energy by adjusting alloy composition rate together with its close or exact lattice matching with GaSb . Investigations in laser diodes and photodetectors utilizing an $\text{AlGa}(\text{As})\text{Sb}/\text{GaSb}$ heterostructure have been carried out by many authors [1-12] as an alternative to an $\text{InGaAsP}/\text{InP}$ system. Although $\text{AlGa}(\text{As})\text{Sb}$ alloy semiconductors have intensively grown by a liquid-phase epitaxial (LPE) technique, problems still remain with the growth of device-quality single crystal and also there still remain unresolved in electrical and optical properties of these epilayers [13]. In order to make the best use of high potential of $\text{AlGa}(\text{As})\text{Sb}/\text{GaSb}$ system as a material for long wavelength optoelectronic devices, it is of great importance to investigate further the growth conditions for high-quality epilayers imperative for a high device

performance.

In this paper, we describe experimental results of LPE-grown AlGaSb or AlGaAsSb on a GaSb substrate. The relationship between the growth temperature and the epilayer properties is investigated by changing the temperature over a wide range from 600 to 400°C. In particular, we demonstrate that the interface instability between the epilayer and the substrate can be effectively suppressed by lowering the growth temperature even in the epilayer with a high Al composition rate. Arsenic addition to ternary layer releases the interface instability: at the same growth temperature, the largest Al composition rate becomes higher in AlGaAsSb quaternary epilayer than in AlGaSb ternary epilayers which can be grown without the instability. Further, it is described that carrier concentration is significantly reduced by decreasing the growth temperature. Finally, it is also reported that two distinct acceptor levels are found to exist in the epilayers by photoluminescence measurements.

2. Experimental procedures.— The epitaxial layers of the ternary or the quaternary alloy semiconductors are grown from Ga-rich solution on (111)-B oriented GaSb substrates using a sliding boat technique in a Pd-diffused hydrogen stream in horizontal furnace. The sliding boat used in this work is composed by a double-bin type crucible which enables us to supply fresh and almost thermal equilibrium melt to a substrate [13]. The GaSb substrate (MCP Electronics LTD.) are both of n-type (Te doped, $n \approx 6 \times 10^{17}$ to $1 \times 10^{18} \text{cm}^{-3}$) and p-type (Zn doped, $p \approx 3 \times 10^{18} \text{cm}^{-3}$). These substrates are etched in a solution of HF:HNO₃:CH₃COOH=1:19:30 (F solution) or Br₂ in methanol and subsequently rinsed in deionized water or methanol, prior to loading to the furnace. The preparation procedures for the substrate are important for growing uniform and smooth epilayers. The substrate orientation must be accurate within $\pm 0.2^\circ$ to obtain an as-grown mirror-like epilayer. The best results are well reproduced with thorough rinse by pure methanol after a final etching of the substrate in the F solution. The raw materials for the melt are 7N-Ga, 5N-Al, 6N-Sb, and a GaAs polycrystal. The dew point at the downstream of the growth tube is lower than -76°C during the growth runs. The heating cycles for the growth are schematically illustrated in Fig. 1. The cycle A is for the melt baking of Ga and Sb, where the temperature is kept at 700°C for 3 to 24 hrs. After the baking, Al and GaAs are added to the melt and the substrate is loaded to the boat (cycle B). For a high temperature growth ($T_G > 500^\circ\text{C}$), the growth solution is heated to a temperature slightly above the growth temperature and maintained at this temperature for 30 min to equilibrate the melt, and then the growth is started (Fig. 1 (a)). For a low temperature growth ($T_G < 500^\circ\text{C}$), a heat cleaning cycle C for the substrate at 550°C for 1 hr has to be added before the growth in order to obtain smooth and uniform alloy epilayers (Fig. 1 (b)). A cooling rate of 0.2 to $0.3^\circ\text{C}/\text{min}$ is used and the thickness of the epilayers is about 0.5 to $15 \mu\text{m}$ depending on the growth interval.

3. Results and discussions.

3.1 Relationship between epilayer growth and substrate preparation process.

In LPE technique, good "wetting" between a melt and a substrate is needed for uniform and smooth morphology together with thickness control of a grown layer. The "wetting" is greatly dependent on a substrate surface condition just prior to the growth.

We investigate the influence of substrate preparation condition and growth temperature on the "wetting" or the growth rate of AlGaSb ternary alloy epilayer.

As shown in Table I, the apparent

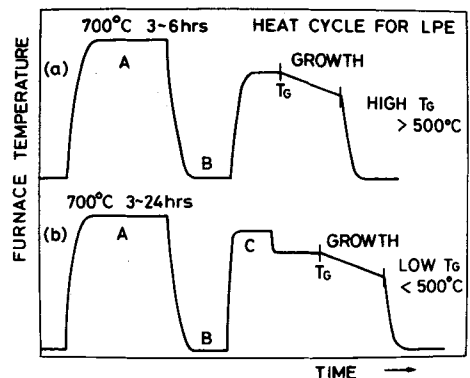


Fig. 1. Temperature profiles of high temperature growth (a) and low temperature growth (b).

growth rate for $\text{Al}_{0.39}\text{Ga}_{0.61}\text{Sb}$ grown at $T_G=500^\circ\text{C}$ exhibited remarkable dependence on the GaSb substrate preparation process. The growth rate is seen to be reduced with decreasing the resistivity of the water used as a final rinse solution for the substrate. At $T_G=400$ and 450°C , $\text{Al}_{0.39}\text{Ga}_{0.61}\text{Sb}$ was not grown at all unless the heat cycle C mentioned in the previous section was introduced prior to the growth regardless of the process subjected to the substrate, whereas no significant difference in the growth rate for $\text{Al}_{0.39}\text{Ga}_{0.61}\text{Sb}$ was observed at a higher T_G than 550°C with various kinds of the final rinse for GaSb.

Judging from the above experimental observation, the surface of the GaSb substrate would be presumably covered by some undesirable thin layer of which thickness is dependent on the quality of the water, which gives great influence on the "wetting" or the apparent growth rate of the epilayer. Although it is still unrevealed whether the thin surface layer covering the substrate surface is formed by an oxide or other materials, the fact that the growth rate at a high T_G is less sensitive to the final rinse solutions for GaSb used in this experiment suggests that the thin surface layer can easily be removed by thermal etching at a high temperature just before the melt comes into contact with the substrate, which leads to a good "wetting" of the melt with a substrate. However, the surface morphology of the epilayer was uniform and smooth when using pure methanol or water with $\rho > 15\text{M}\Omega\cdot\text{cm}$ as a final rinse solution for the substrate. Thus, the thickness control of the epilayer with excellent morphology becomes easier in the LPE growth at a lower temperature.

3.2 Low-temperature growth of AlGaSb and AlGaAsSb

The low-temperature growth of AlGa(As)Sb has been found to be very effective to reduce the concentration of background impurities or defects[9, 13, 14] and further to suppress the interface instability between the alloy epilayer and the GaSb substrate[13]. Calculations and experiments for the phase diagram of the ternary alloy semiconductors have been carried out by several authors[15-19]. However, there are only a few reports [13, 14, 17] on experimental studies for low-temperature LPE growth

Table I Relationship between substrate preparation processes and $\text{Al}_{0.39}\text{Ga}_{0.61}\text{Sb}$ growth characteristics.

Final rinse solution	Thickness (μm) of the epilayer at $T_G = 500^\circ\text{C}$	Surface morphology
(a) Pure methanol	9.3	mirror-like
(b) Water ($\rho > 15\text{M}\Omega\cdot\text{cm}$)	7.5	mirror-like
(c) Water ($\rho \sim 1\text{M}\Omega\cdot\text{cm}$)	2.8	rough surface
(d) Water ($\rho \sim 3\text{K}\Omega\cdot\text{cm}$)	0~2.9	no growth or rough surface

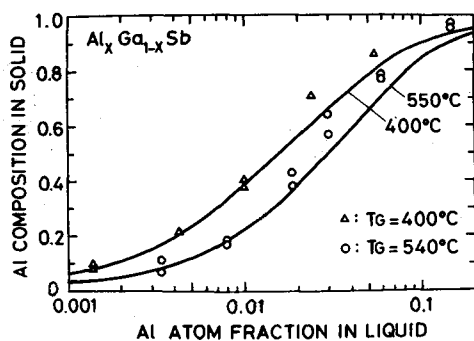


Fig. 2. Solidus isotherms in AlGaSb at $T_G=400$ and 540°C . Solid curves are the calculated results after Cheng et al.[17]

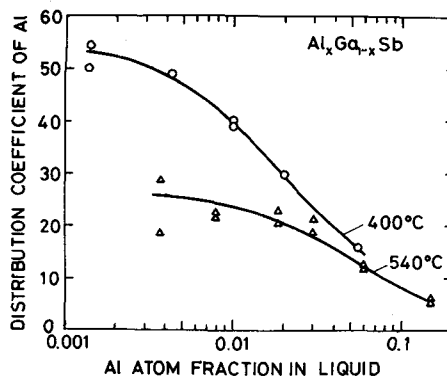


Fig. 3. Al distribution coefficient of AlGaSb at $T_G=400$ and 540°C .

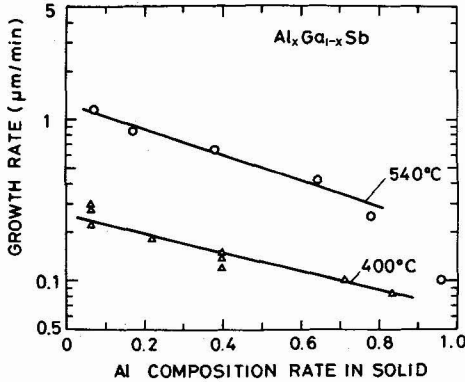


Fig. 4. Relationship between AlGaSb growth rate and Al composition rate in solid.

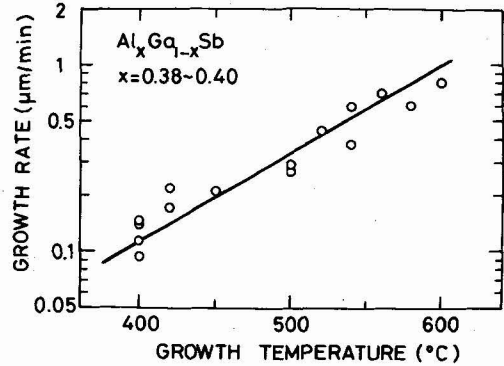


Fig. 5. Growth temperature dependence on the growth rate of AlGaSb .

th of these ternary and quaternary alloy semiconductors. In order to obtain high-quality heteroepitaxial layers of these materials, the LPE growth was performed at T_G 's below 540°C .

Experimentally determined solidus isotherms in AlGaSb system grown at 400 and 540°C , are shown in Fig. 2. These experimental results are in good agreement at a low X_{Al} region with the calculated results based on a simple solution model reported by Cheng et al.[17]. Figure 3 shows the relationship between Al atom-fraction in solid phase and in liquidus phase, i.e., Al distribution coefficient. It is seen that Al distribution coefficient, $X_{\text{Al}}^s/X_{\text{Al}}^l$, in AlGaSb at $T_G=400^\circ\text{C}$ decreases from 50 to 15 with increasing X_{Al} from 0.001 to 0.55. The raising of T_G to 540°C reduces $X_{\text{Al}}^s/X_{\text{Al}}^l$ by approximately one half within X_{Al} employed in this work. The growth rate decreases with increasing X_{Al} at both T_G 's, as shown in Fig. 4. In particular, a submicron AlGaSb epilayer can be easily reproduced by decreasing T_G to 400°C . Figure 5 indicates the growth temperature dependence on the growth rate of $\text{Al}_x\text{Ga}_{1-x}\text{Sb}$ with $x=0.38 - 0.40$.

It is well known that crystal instability is often observed at the interface in a heteroepitaxial growth of AlGaSb on GaSb [13, 17, 20, 21]. We observed that the low-temperature growth of AlGaSb is very effective to suppress the interface instability and that the As addition to the melt, i.e., the quaternary epilayer growth leads to uniform straight interface. The Al composition rate x in $\text{Al}_x\text{Ga}_{1-x}\text{Sb}$ beyond which the interface becomes nonuniform was extended from 0.47 to 0.84 by lowering T_G from 600 to 400°C . Figure 6 shows a typical example of $\text{Al}_{0.84}\text{Ga}_{0.16}\text{Sb}$ layer surface and cleaved cross section where T_G is 400°C . No interface instability is observed. Relationship between the interface instability and the growth temperature as well as the As addition is shown in Fig. 7 where one can clearly see that the instability is able to be suppress by lowering T_G or by the As addition even in the heteroepitaxial growth of these alloys with a high Al composition rate. The avoidance of instability is essentially important in wide-gap heterojunction device applications. In general, interface instability is caused by a constitutional supercooling phenomenon[22].

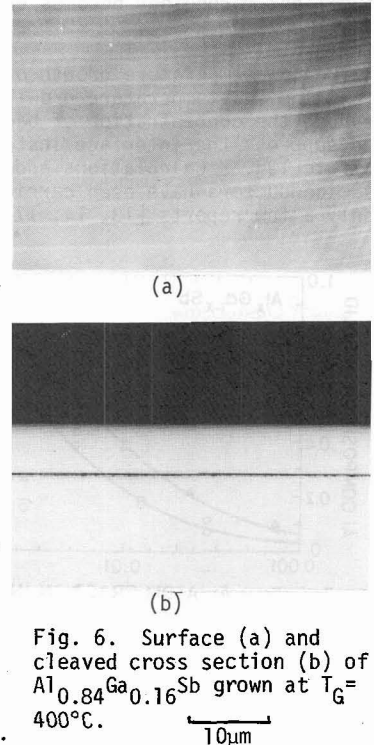


Fig. 6. Surface (a) and cleaved cross section (b) of $\text{Al}_{0.84}\text{Ga}_{0.16}\text{Sb}$ grown at $T_G=400^\circ\text{C}$.

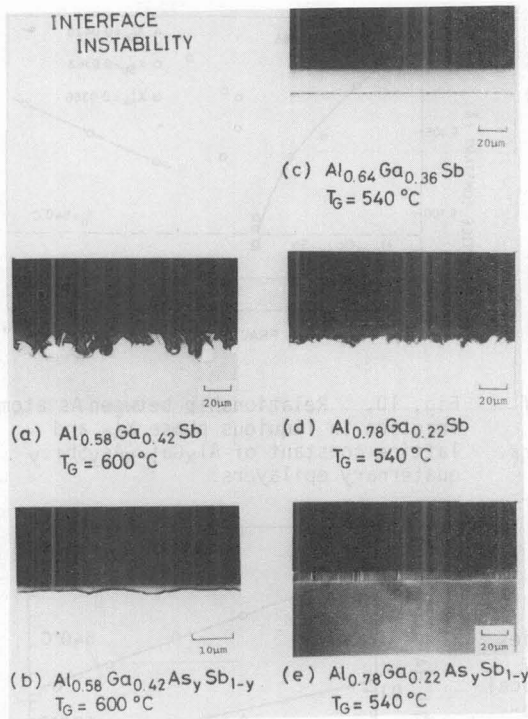


Fig. 7. Relationship between interface instability and growth temperature.

constitutional supercooling. At the same T_G , on the other hand, the growth rate decreases with increasing Al composition rate as shown in Fig. 4. However, the instability is easy to occur in the growth at higher Al composition rates. Therefore, it cannot be explained only in terms of the growth rate. Since the critical value of G_L/v depends on the liquidus slope, the liquid concentration, the distribution coefficient and the diffusion constant at the interface of each component, it is further required to take these factors into account for the interpretation of the instability phenomenon.

The As composition rate is quite limited in the AlGaAsSb quaternary alloy system, which results in difficulty in growing AlGaAsSb lattice-matched with GaSb. This would be due to low solubility of As into the liquidus phase[2] or due to a miscibility gap in a Ga-As-Sb system[23, 24]. Therefore, AlGaAsSb with a high As-composition lattice-matched with GaSb cannot be grown at a low growth temperature[9, 19]. However, even in the quaternary epilayer lattice-mismatched with GaSb, we were able to reproducibly grow the excellent morphological epilayer with a high Al composition rate, which is free from the interface instability as shown in Fig. 8.

Figure 9 shows the T_G dependence of the As composition rate in solidus phase of AlGaAsSb. At $T_G=400^\circ\text{C}$, the maximum As composition rate y in $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}_y\text{Sb}_{1-y}$ is estimated around 0.0045. $\text{Al}_{0.39}\text{Ga}_{0.61}\text{As}_y\text{Sb}_{1-y}$ with $y=0.025\sim 0.03$ lattice-matched with $\text{Al}_{0.07}\text{Ga}_{0.93}\text{Sb}$ was reproduced by raising T_G up to 540°C as shown in Fig. 10. However, the increase in As atom fraction in liquidus phase leads to irregular variation in the lattice constant of the quaternary layers, which is probably caused by the miscibility gap of this quaternary system[2, 24]. Since the growth rate of the quaternary epilayer is significantly decreased with the As composition rate in liquidus phase as indicated in Fig. 11, we can successfully reproduce the submicron ($<0.5\ \mu\text{m}$) quaternary layers, which is very useful for double heterojunction applications.

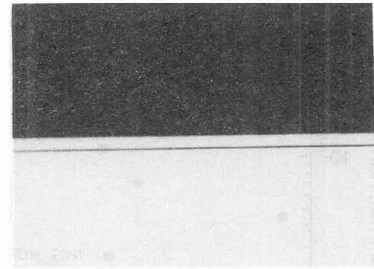


Fig. 8. Cleaved cross section of $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}_y\text{Sb}_{1-y}$ with $y=0.007$ grown at 540°C on GaSb.
20µm

Assume that the instability in the present experiments is due to the supercooling phenomenon.

In order to avoid the supercooling, a criterion must be followed that required G_L/v be greater than a certain value[22], where G_L is the temperature gradient in the liquidus at the interface and v the growth velocity. If G_L is assumed to be constant in each growth run in Fig.7, a value of G_L/v would be increased with reducing T_G or with the As addition since the growth rate is significantly decreased by lowering T_G or by the As addition, which agrees with the requirement for suppression of

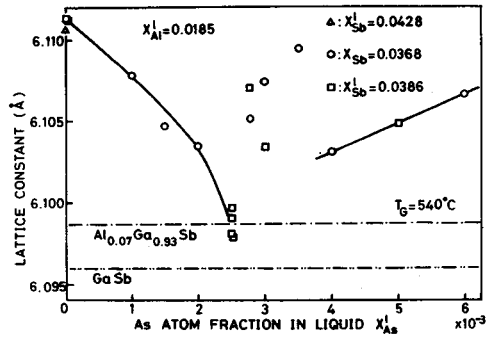
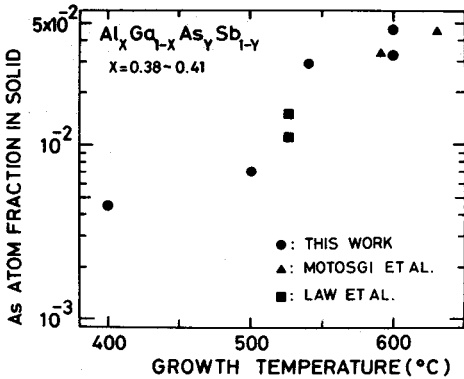


Fig. 9. Growth temperature dependence of the As composition rate in solidus phase in $Al_{0.4}Ga_{0.6}As_ySb_{1-y}$ quaternary epilayers. The results by Motosugi et al.[19], and Law et al.[2] are also indicated for comparison.

Fig. 10. Relationship between As atom fraction in liquidus phase X^I_{As} and lattice constant of $Al_xGa_{1-x}As_ySb_{1-y}$ quaternary epilayers.

3.3 Electrical and luminescent properties of $AlGaSb$.

In order to investigate the electrical properties of epilayers, the Hall measurements have been carried out by a p-n junction separation technique because of a semi-insulating $GaSb$ substrate unavailable.

The undoped $AlGaSb$ epilayers show p-type conduction regardless of T_G 's employed at present work. However, hole concentrations are significantly reduced by about two orders in magnitude by lowering T_G from 600 to 400 °C as shown in Fig. 12. $Al_{0.4}Ga_{0.6}Sb$ epilayer at $T_G=400^\circ C$ is seen to have a typical value of $p=8 \times 10^{15} cm^{-3}$ at room temperature. Temperature dependence of hole concentration of the epilayers at $T_G=400$ and $536^\circ C$ is shown in Fig. 13. These results could allow us to evaluate acceptor levels, the acceptor and the compensating donor concentrations by computer analyses. Although attempts to estimate these material parameters of the epilayers by the computer analyses have been made, it is found to difficult to evaluate the reasonable values of the parameters because of remarkable influence on the Hall measurements at a high temperature region of leakage current of p-n junction used electrical separation between the epilayers and the substrates. A rough estimation from the data at a low temperature region gives a value of 25 to 30 meV for an acceptor level.

In undoped p-type $GaSb$, a model of doubly-ionizable acceptor whose origin is connected with an intrinsic Sb vacancy has been reported[25]. It has been indicated, on the other hand, that by reducing T_G undoped $AlGaSb$ or $GaSb$ epilayers exhibit n-type[9, 14, 26] and further residual donor concentration decreases drastically up to $2-4 \times 10^{15} cm^{-3}$ for $Al_{0.2}Ga_{0.8}Sb$ and up to $2 \times 10^{13} cm^{-3}$ for $GaSb$ [14, 26]. The origin of p-type conduction in our epilayers is still unknown whether it is due to intrinsic defects related to a Sb vacancy[25] or to extrinsic impurities such as Si [30]. Further experiments for obtaining high-purity epilayers are needed to elucidate the origin.

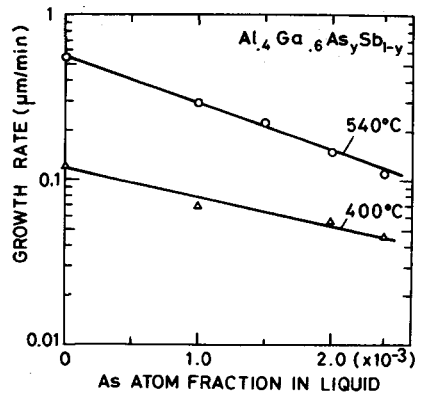


Fig. 11. Dependence of growth rate of $Al_{0.4}Ga_{0.6}As_ySb_{1-y}$ quaternary epilayers on the As atom fraction in liquidus phase X^I_{As} .

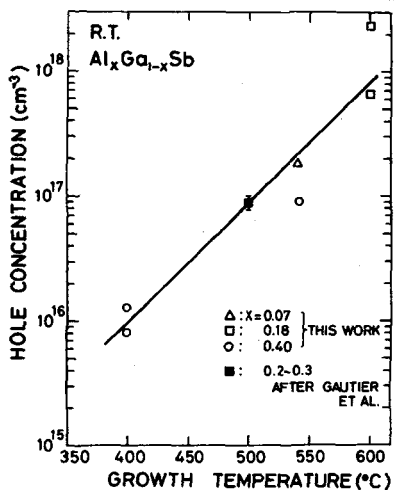


Fig. 12 Growth temperature dependence of hole concentration of p-AlGaSb layer. Solid square is after Gautier et al.[32].

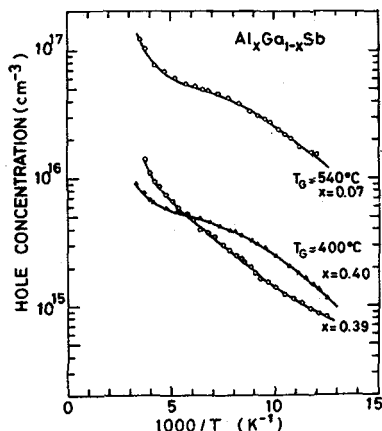


Fig. 13. Temperature dependence of hole concentration of p-AlGaSb at $T_G=400$ and 540°C . Rapid increase in hole concentration is due to leakage current from p-n junction at high temperatures.

Photoluminescence (PL) measurements were made for $\text{Al}_{0.07}\text{Ga}_{0.93}\text{Sb}$ ternary epilayers grown at T_G 's from 550 to 400°C to obtain information on impurity or defect levels involved. A high-pressure Hg lamp with a filter or argon ion laser (1W) for excitation source and a cooled PbS photoconductive cell for a detector were used for the measurements.

Fig. 14 shows PL spectra measured at 77 and 4.2K in an epilayer grown at $T_G=450^\circ\text{C}$. At 4.2K four emission bands labelled as I_x , I_1 , I_a , and I_c can be observed at 0.873, 0.870, 0.844, and 0.815eV, respectively, while one can see two distinct emission bands of I_0 and I_a' located at 0.870 and 0.842eV at 77K, respectively.

The I_0 emission band at 77K varies in peak photon energy depending on the slight difference in the Al composition rate among the epilayers grown at different T_G 's, as shown in Fig. 15. However, the peak energy difference between I_0 and I_a' remains almost unchanged among the epilayers: the value is about 27meV. Further, the intensity of I_a' with respect to that of I_0 tends to decrease at lower T_G 's which lead to decrease in the acceptor concentration.

From these results, the I_0 emission band may be arisen from direct transition from electron in the conduction band to hole in the valence band, deducing from data of temperature dependence of bandgap for GaSb together with Al composition dependence of that for the ternary alloy system[28-30]. And in addition, I_a' can be ascribed to radiative recombination due to a transition of free electron into an acceptor level E_A located at about 27meV above the valence band. This value is interpreted with the acceptor level deduced from the electrical measurements.

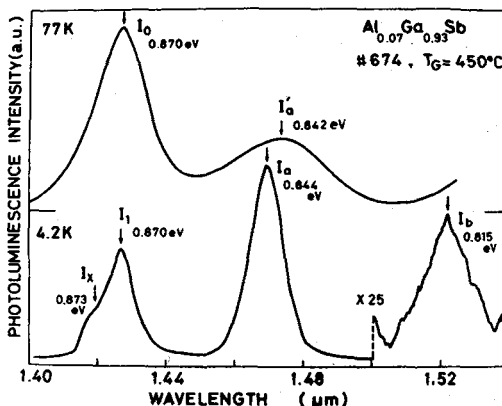


Fig. 14. PL spectra at 77 and 4.2K of an $\text{Al}_{0.07}\text{Ga}_{0.93}\text{Sb}$ epilayer grown at 450°C .

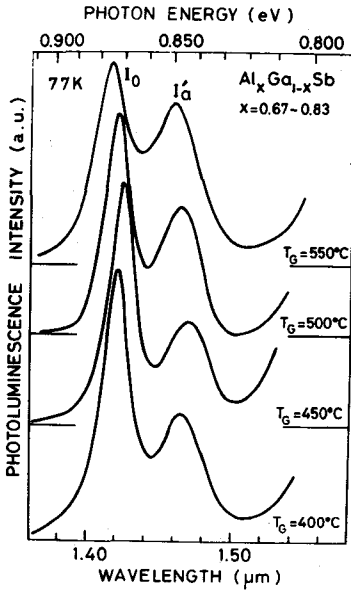


Fig. 15. PL spectra at 77K of $Al_xGa_{1-x}Sb$ grown at various temperatures.

Table II Summary of emission peak energies and proposed mechanisms for PL spectra of $Al_xGa_{1-x}Sb$ with $x=0.07$.

4.2K Emission band	Peak energy (eV)	Mechanism	77K Emission band	peak energy (eV)	Mechanism
I_x	0.873	Free exciton	I_0	0.870	direct band to band
I_1	0.870	Exciton bound to neutral acceptor	I'_a	0.842	Free to shallow acceptor
I_a	0.844	D-A pair(donor-shallow acceptor)			
I_b	0.815	D-A pair(donor-deep acceptor?)			

At 4.2K, the emission intensity of I_1 band increases strongly with increasing the excitation intensity but no displacement of the emission peak energy is observed, and in addition, the emission intensity of I_1 is strongly dependent on T_G : the I_1 intensity of an epilayer grown at $T_G=400^\circ C$ is relatively stronger than that of an epilayer at $T_G=550^\circ C$ whose carrier concentration is higher by about one order in magnitude than that grown at $T_G=400^\circ C$. Further, the I_1 intensity is drastically reduced at above 4.2K. These observations strongly suggest that the I_1 band is due to radiative recombination from exciton bound to neutral

acceptor, considering that the epilayers are p-type. Similar carrier concentration dependence on emission intensity of bound exciton complex to our results has been observed in GaAs[31]. The relatively broad emission band of I_1 would be ascribed to fluctuation of energy state due to perturbed disorder arrangement of the constituent group-III atoms in the lattice.

The binding energy of free exciton, E_{ex} for $Al_{0.07}Ga_{0.93}Sb$ can be estimated to be $E_{ex}=3meV$ by extrapolating physical constants for each binary compound[23]. If the highest energy emission band I_x located at 0.873eV is assumed to be due to free exciton emission, the band gap energy E_G at 4.2K for the alloy epilayer is evaluated to be 0.876eV which is smaller by about 15meV than that deduced from PL data reported by Allegre et al.[28]. This relatively large discrepancy in the energy gap is not clear, but it would be caused by the possibility that the Al composition rate determined by an X-ray diffraction technique slightly differs from the rate of the actual epilayer. Using E_A , E_{ex} , and E_G thus estimated, we can calculate the binding energy of I_1 . From an effective mass arguments developed by Hopfield[27], the photon energy emitted by radiative recombination of exciton bound neutral acceptor, $E(A^0,X)$ is given by a following equation: $E(A^0,X) = E_G - E_{ex} - 0.06E_A$ where the electron mass, m_e and the hole mass, m_h for $Al_{0.07}Ga_{0.93}Sb$ is taken as $0.0476m_0$ and $0.479m_0$ [23] (m_0 is free electron mass), respectively. Then, the calculated value for $E(A_0,X)$ is 0.871eV which is in good agreement with the peak energy of I_1 .

As for I_a observed at 4.2K, we can assign it as the D-A pair emission because the emission peak position is slightly displaced to higher energy as the excitation intensity increases. Although the emission intensities of I_b among the samples is too weak to characterize the emission mechanism, it is presumably arisen from the D-A pair emission where the related acceptor is deep one with an activation energy of around 50 to 60meV. Table II shows the summary of the peak energies and the proposed mechanisms of each line observed by the PL measurements.

4. Conclusions. - The experimental results obtained in this work can be summarized in the following:

- (i) The morphology and the growth rate of AlGa(As)Sb epilayers are greatly affected by the GaSb substrate preparation processes when growing at a low temperature of 400 or 450°C. A heat cleaning process at 500 or 550°C just prior to the low temperature growth gives the excellent epilayers.
- (ii) Interface instability can be avoided by lowering T_G . At $T_G=540^\circ\text{C}$, the largest Al-composition rate for a ternary layer without the instability is around 0.64, whereas it for a quaternary layer extends to about 0.78.
- (iii) Lowering the growth temperature contributes significantly to reduction in carrier concentration: $p=10^{18}\text{cm}^{-3}$ at $T_G=600^\circ\text{C}$ whereas $p=8\times 10^{15}\text{cm}^{-3}$ at $T_G=400^\circ\text{C}$.
- (iv) PL spectra of $\text{Al}_{0.07}\text{Ga}_{0.93}\text{Sb}$ show four emission bands at 4.2K. I_x at 0.873eV, I_1 at 0.870eV, I_a at 0.844eV, and I_b at 0.815eV are presumably arisen from free exciton, exciton bound to neutral acceptor, donor-shallow acceptor pair, and donor-deep acceptor pair, respectively. At 77K, band-to-band and free-to-bound recombination are dominant in the ternary epilayer.

Experiments will be further continued to obtain the high-quality epilayers imperative for optoelectronic device applications, which also enables us to elucidate bulk and surface properties of AlGa(As)Sb alloy semiconductors

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