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A Study of High-Quality InAs-Strained Bulk Layer Growth on GaAs by MOCVD

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ABSTRACT

The interface between GaAs and InAs layers possesses good hetero-structure epitaxial growth by using MOCVD (metal organic chemical vapor deposition) method. The main peak of an InAs X-ray is sharp, the full-width half-maximum (FWHM) being narrow for the V/III ratio near 75, at only about 0.342°. A Laue pattern and a scanning electron microscopy image both show the InAs layer grown on a GaAs layer with a single crystalline structure. The *i*-GaAs buffer layer is grown on a (100) semi-insulated GaAs substrate at 600°C at a mole fraction ratio of group V to group III with an optimum of 100. The growth rate of the GaAs layer is 0.11 *μm/min*. The *n*-like InAs layer formed as a high-strained layer is also grown on the GaAs buffer layer at a substrate temperature of 600°C by a V/III mole fraction ratio with an optimum of 75. Consequently, the growth time of the InAs layer cannot exceed 20 minutes. The growth rate of the InAs layer is about 0.12 *μm/min* at a growth temperature of 600°C. A carrier concentration of $2\times10^{17}/cm^3$ and a Hall mobility of $2\times10^3 cm^2/V$-sec were obtained for an undopant *n*-like InAs prepared on a GaAs substrate at a growth temperature of 600°C at a group V/III ratio of 75.

Key Words: InAs, hetero-structure, MOCVD, strained layer

高品質砷化銦在砷化鎵基板上成長之研究

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摘　要

砷化銦成長在砷化鎵基板上經由掃描式電子顯微鏡及勞厄圖像顯示具有單晶品質及良好異質結構介面。使用有機氣相磊晶法成長砷化銦，經由 *x* 光檢測 75 莫耳比、600°C 成長溫度下的砷化銦磊晶層其半高寬僅達 0.342°。*n*-like 砷化銦磊晶層在 75 莫耳比與 600°C 成長溫度下具有 $2\times10^{17}/cm^3$ 的載子濃度與 $2\times10^3 cm^2/V$-sec 的載子移動率。本質砷化鎵緩衝層在 600°C 成長溫度下可以藉由有機氣相磊晶法在三五族莫爾比最佳化條件下成功的成長在（100）半絕緣砷化鎵基板上，三五族莫耳比最佳化條件約為 100。在最佳化三五族莫耳比條件下砷化鎵成長速率約為每分鐘 0.11 微米。類似 *n* 型的砷化銦薄膜亦在 600°C 成長溫度下成功的成長在與其形成高應力介面之砷化鎵緩衝層上，三五族莫耳比最佳化條件亦約為 75。砷化銦薄膜在砷

化鎵緩衝層上之成長速率約爲每分鐘 0.12 微米，但是成長厚度不能超過 2.4 微米，否則將產生明顯的缺陷。雖然砷化銦／砷化鎵應力量子井會造成能階變化，但是砷化銦／砷化鎵量子井應用於光電元件的價值性高，本研究利用有機氣相磊晶法高速成長出薄膜厚度可控性高、介面品質可控性高、摻雜可控性顯著之砷化銦／砷化鎵異質介面，因此該技術將可以應用於雷射元件製造業之生產。

關鍵詞：砷化銦，異質結構，有機氣相磊晶法，應力層

I. INTRODUCTION

The GaAs material is belong to zincblende structure [10, 12] and the active energy EA of Ga and As in GaAs material are 6.0ev and 3.2ev, respectively [16-17]. The lattice constant of GaAs material is about 5.65325Å at 300ºK [10, 12] and the energy band type of GaAs belong to direct band gap [16]. The GaAs material possessed light effective mass, so the electron mobility is larger [11]. So as to GaAs material can be applied in higher frequency devices as like resonant tunneling diodes (RTD) [9, 23-24] photodetectors [3, 18], hetero-structure bipolar transistor (HBT) [6, 14], high electron mobility transistor (HEMT) [15, 22], and Laser diodes [13, 19] etc. The InAs material belonged to zincblende structure also. The lattice constant of InAs material is about 6.0584Å at 300ºK, and the energy band type of InAs compound is direct band structure [16-17]. The InAs material possessed not only wider energy band gap but also much higher electron mobility about 30,000 cm^2/V·S and lower LO phonon about 29.6 mev at room temperature than other III-V semiconductor compounds [10, 12]. The compound of GaAs and InAs semiconductor called $In_x Ga_{1-x}As$ combined the all merits of GaAs material and InAs material characteristics, so the $In_xGa_{1-x}As$/GaAs devices have been comprehensively grown and applied recently [6, 13-15, 18-19, 22]. Because the difference of energy band (△Eg) between InGaAs and GaAs material is smaller than that between InAs and GaAs material about 1.06 ev, we studied the InAs material grown on the GaAs material formed type I structure.

InAs quantum wells on GaAs substrates have attracted much attention for potential applications to optoelectronic devices as well as for fundamental study of quantum effects. It has been recently shown that quantum wells can be obtained from a strained InAs layer directly grown on a GaAs substrate with molecular beam epitaxy (MBE) [5, 7, 20] and organomtetallic vapor phase epitaxy (OMVPE) [4, 21]. The growth surface containing the lattice-mismatched material islands, compensates an increase in energy due the extra free surface wuth a decrease in strain-energy for the growth methods. These techniques must use the strain-induced islanding that may be result in a increase of epitaxy cost.

The higher barrier height of InAs/GaAs hetero-structure will result in excellent devices characteristic perfectly, if the strain effect of InAs/GaAs heterojunction can be overcome. The functions of InAs/GaAs quantum well or superlattice devices will much better. We studied the bulk characteristics of InAs/GaAs high lattice mismatch here by using metal organic chemical vapor deposition growth (MOCVD) method. MOCVD can be used to precisely control the thickness and chemical composition of heteroepitaxial layers, as well as to introduce dopant species in-stiu. The thickness and interface control available in current MOCVD has produced a mature quantum well technology, and a rich array of semiconductor physics and devices for semiconductor industry in Taiwan.

II. EXPERIMENTAL SYSTEM AND PROCEDURES

The InAs/GaAs hetero-structure is grown by MOCVD system. This system including the reaction chamber system, the exhausted system, the gas handling system, and the automatic pressure control (APC) system. The reactor is designed by horizontal type chamber constructed with a rectangular cross section (2.5×2 cm^2) and a water cooling jacket on the top of growth zone. The susceptor was made of Mo with an area of 5×2 cm^2 and 5º titled angle which can provide uniform growth rate crossing the substrate. The exhausted system is composted by the one turbo pump and two rotary pumps. The two rotary pumps are connected with main line system and bypass line system, respectively. The turbo pump is connected with main line system that can keep the pressure of reactor, in order to improve the growth quality of the film. In the gas handling system, under 10^{-3} Torr the palladium-diffused H_2 was used as the carrier gas and the purge gas. The H_2 is send to the reactor system after H_2 is purified by 0.5 μm particle filter, the removing oxygen, the moister filter, and the H_2 purifier containing palladium at the operating temperature range of 370°C to 400°C. The automatic pressure control system can stabilize the pressure of growth system during the growth process. It consists of a pressure sensor, a pressure display, a pressure controller, and an exhaust throttle valve.

The growth temperature and pressure of InAs/GaAs is about at 600°C and 150 Torr, respectively. Thriethylgallium (TEGa) and trimethylindium (TMIn) bubblers were maintained at 10°C and 20°C, respectively. The concentration of arsine (AsH_3) with hydrogen is 15.1%. In this work, we studied the characteristics of InAs/GaAs bulk material by changing the V/III mole fraction ratio of InAs material and the growth time of InAs layer.

The surface morphology of InAs bulk layer is viewed by optical electron microscopy for different V/III mole fraction ratios and growth times of InAs material. The growth rate of InAs bulk layer on GaAs buffer layer is measured by scanning electronic microscopy from the cross section of InAs layer. The x-ray diffraction can see the quality of InAs layer by judge of full width at half maximum (FWHM). The quality of InAs layer grown on GaAs material can be also understood by x-ray diffraction Laue pattern.

III. RESULTS AND DISCUSSIONS

The mole fraction ratio of group V and group III so called V/III ratio is important, for growth InAs strained layer on the GaAs layer. We calculated the V/III ratio, as follow

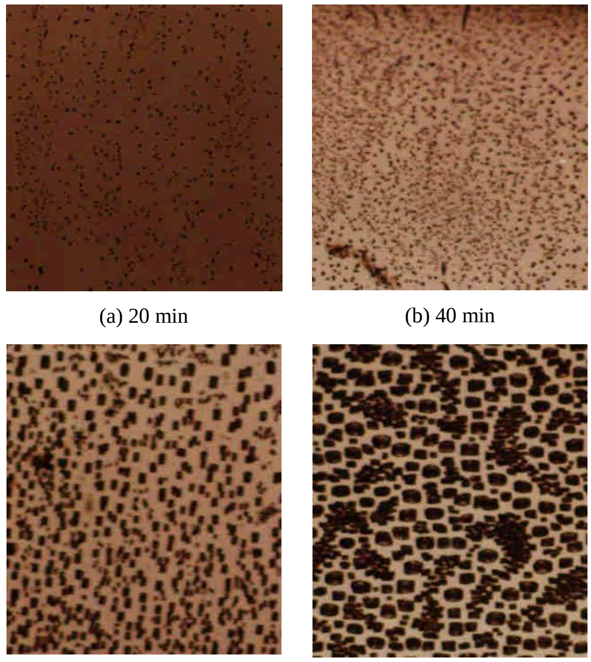
$$R = \frac{M_V}{M_{III}} \tag{1}$$

where R is V/III mole fraction ratio, M_{III} is mole value of group III, and M_V is mole value of group V.

1. Optical Electron Microscopy

The surface morphology of InAs layer grown on GaAs material is viewed by optical electron microscopy in multiplication of 500 times. We can study the different surface morphology of InAs layer at the various growth parameters. The strained effect and optimum growth conditions will be able to understand slightly for InAs compound grown on GaAs layer by optical electron microscopy.

The GaAs buffer layers are grown by V/III ratio of 100 optimum value at substrate temperature of 600°C in 30 minutes for all InAs/GaAs hetero-structure. The surface morphology of InAs structures layer grown on GaAs buffer layer at substrate temperature of 600°C by V/III ratio of 75 in every grown time is shown in Fig. 1. Making a comparison with Fig. 1(a), 1(b), 1(c), and 1(d), we can understand that when thickness of InAs strained layer is thick in longer growth time, the surface morphology of InAs is found rough. Because of



(a) 20 min (b) 40 min

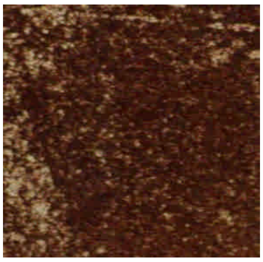
(c) 1 hr (d) 1 hr. 40 min

Fig. 1. The surface morphology of InAs strain layer

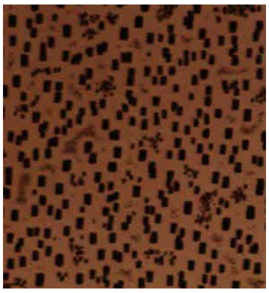
dislocation defects of InAs/GaAs interface resulting in strained effect, the dangling bond of InAs and vacancy defects result in the impurity autodoping in the institute defect for weak As-ionic bond. The thicker the strained layer is, the more serious the phenomenon is. So the optimum growth time of InAs strained layer grown on GaAs substrate at growth temperature of 600°C is less than 20 minute. This result can be fit by growth rate of InAs and the relationship between critical thickness L_C and lattice constant a of InAs given [1] by

$$L_C = \frac{a-(1-\frac{\sigma_p}{4})}{0.144\sqrt{2}\pi(1+\sigma_p)}[\ln\frac{\sqrt{2}L_C}{a}+1] \tag{2}$$

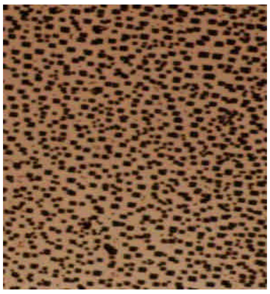
where σ_P is the Poisson's ratio which usually equals to 1/3. In Fig. 2, the surface morphology of InAs layer is shown for different V/III ratio value at the same buffer layer growth condition. The GaAs buffer layer are grown by V/III ratio of 50 and in 30 minute at substrate temperature of 600°C. The InAs strained layer grown on GaAs buffer layer are grown by three kind V/III ratio and all in one hour at growth temperature of 600°C. The surface morphology is rough when the V/III ratio is less than 5, due to the Ga-rich resulting in vacancy defect. The surface morphology is smooth and have a little black spots when the V/III ratio is larger than 75, due to the As-rich slightly resulting in institute defect. So the optimum V/III ratio value of growth InAs strained layer is about 75.



(a) V/III ratio of 5

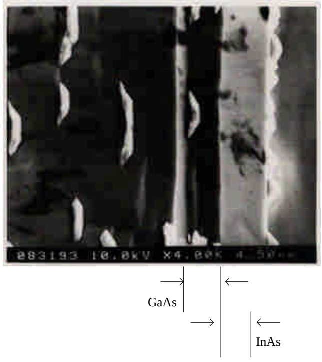


(b) V/III ratio of 50

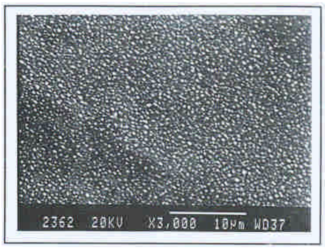


(c) V/III ratio of 75

Fig. 2. The surface morphology of InAs strain layer



(a) The cross section of InAs/GaAs structure



(b) Surface morphologies of InAs epilayer grown on GaAs

Fig. 3. SEM photograph of InAs/GaAs structure

2. Scanning Electron Microscopy (SEM)

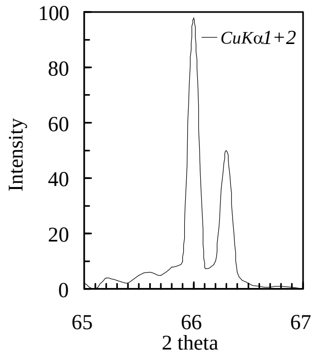
Fig. 3 is clearly appeared the cross section of InAs/GaAs structure by scanning electron microscopy without using chemical etching. The structure is fabricated by growth GaAs buffer layer on (100) semi-insulated GaAs substrate at substrate temperature of 600°C in V/III ratio of 50 firstly. Then the InAs compound is grown by V/III ratio of 75 at growth temperature of 600°C on GaAs buffer layer that already grown on GaAs substrate before. From the cross section photography of InAs/GaAs hetero-structure by scanning electron microscopy of multiplication of 1000 times, we can found the thickness of GaAs buffer layer about 2.2μm in growth time of 30 minute and of InAs strained layer about 2.392μm in growth time of 20 minute. According to the growth dynamic mechanism, the experimental values of growth thickness of InAs strained layer and GaAs buffer layer are correspond to the calculated. From the Fig. 3(a), the growth rates of GaAs buffer layer grown on GaAs substrate and InAs strained layer grown on GaAs buffer layer are about 110Å/mm and 119Å/mm, respectively. Although the InAs material possessed larger lattice mismatch than GaAs material grown on GaAs substrate, the growth rate of InAs strained layer on GaAs layer is larger than that of GaAs buffer layer on GaAs substrate at the same growth conditions. The reason is that the active energy of In-source is larger than that of Ga-source with As-source. Fig. 3(b) shows the SEM photograph of surface morphologies of InAs epilayer grown on GaAs substrate at 600°C.

3. x-ray Diffraction

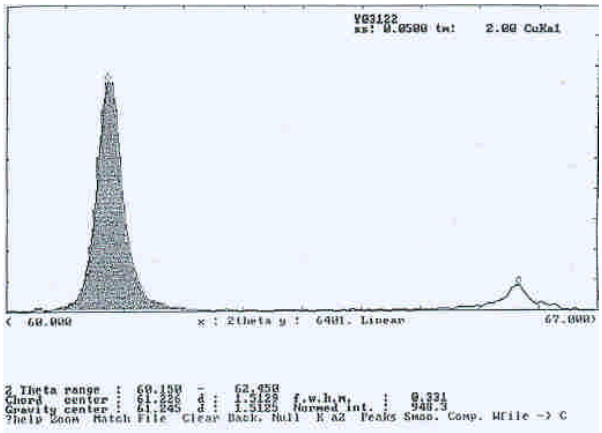
The x-ray diffraction of InAs layer is measured by Simens-XRD instrument. The x-ray diffractions of GaAs buffer layer are shown in Fig. 4(a). The x-ray diffraction of InAs strained layer at growth temperature of 600°C by V/III ratio of 75 shown in Fig. 4(b). In Table 1, making comparison with the main peaks of x-ray diffraction, the main peak is larger at about V/III ratio of 100, because in this V/III ratio the GaAs layer grown on GaAs substrate does not result in Ga-rich or As-rich. The main peak is sharp and the full width half maximum is narrow for the V/III ratio near 100 for GaAs layer about 0.0421°. The shift of 2 theta (2θ) is also smaller for the V/III ratio near 100. In Table 2, it made comparison with the main peaks of x-ray diffraction for InAs layers. The main peak is sharp and the full width half maximum is narrow for the V/III ratio near 75 for InAs layer about 0.342°.

4. Laue Pattern of x-ray Diffraction

The InAs/GaAs high lattice mismatch strained structure is grown by using the growth condition that the growth times of *n*-like InAs bulk layer by V/III ratio of 75 and GaAs buffer layer at growth temperature of 600°C by V/III ratio of 100 are 20 minutes and 1 hour, respectively. The Laue pattern of *x*-ray is shown in Fig. 5. It indicated the light spot obviously. So we can prove that InAs compound has been grown on the GaAs buffer layer successfully. The light spot of Laue pattern



(a) The x-ray diffraction of GaAs buffer layer



(b) The x-ray diffraction of InAs strained layer

Fig. 4. The x-ray diffraction on InAs/GaAs structure

Table 1. The x-ray measurement of GaAs buffer layer

GaAs Buffer Layer V/III Mole Fraction Ratio	2 theta (2θ)	$\frac{K\alpha 2}{K\alpha 1}\%$	FWHM
27	65.993°	49.96	0.0474°
100	66.001°	47.44	0.0421°
130	65.997°	49.24	0.0471°
GaAs Substrate	66.020°	47.79	0.0579°

is not so light, due to the larger difference of lattice constant between InAs material and GaAs material. On the neighborhood of InAs/GaAs layer interface region, the atoms array of InAs and GaAs is distortion both, formed a dislocation phenomenon. This will make the refraction light of the x-ray incident light become broaden light spot. Due to only 1μm thickness of InAs thin film, the disorder atom array of InAs/GaAs interface dislocated by strained effect may be shown by this x-ray Laue pattern measurement. The positions of light spots for this Laue pattern is seem that the direction of InAs crystal on the GaAs buffer layer followed the (100)

Table 2. The x-ray measurement of InAs strained layer

InAs Strained Layer V/III Mole Fraction Ratio	2 theta (2θ)	FWHM
5	60.99°	2.67°
18.7	60.99°	1.54°
37	61.01°	0.82°
40	61.21°	0.69°
75	61.23°	0.34°
90	61.22°	0.64°
140	61.22°	1.21°
270	61.21°	2.67°
GaAs Substrate	66.020°	0.0579°

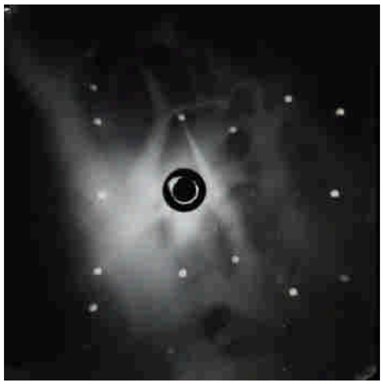


Fig. 5. The x-ray Laue pattern of InAs layer by V/III ratio of 75 in growth time of 20 minutes

direction of GaAs substrate successfully. But due to the high lattice matched for this InAs/GaAs structure and not thick enough for InAs layer, the distance between every spot for this Laue pattern is slightly different with the distance of original InAs compound (100) direction.

5. Strained Effect of InAs/GaAs Hetero-Structure

Due to the lattice mismatch of InAs and GaAs layer is high, the thickness of InAs layer grown on GaAs buffer layer is important. The relation of between the strained effect and band structure for InAs/GaAs strained heterojunction. At first, we calculated the band structure of InAs/GaAs junction influenced by strained effect. There are two terms in the strain Halmiltonian, one is the hydrostatic term, the other is the shear term. The hydrostatic term, ΔE_H, is given by [8]

$$\Delta E_H = 2Q_H \frac{C_{11} - C_{12}}{C_{11}} e_{xx} \qquad (3)$$

where C_{11} and C_{12} that are the elastic constants of InAs are about 8.329×10^{11} and 4.526×10^{11}, respectively. The e_{xx} is the strain tensor about -0.067 depicted by

$$e_{xx} = \frac{a_{GaAs} - a_{InAs}}{a_{InAs}} \quad (4)$$

where a_{GaAs} and a_{InAs} that are the lattice constants of GaAs and InAs are about 5.6533Å and 6.0584Å, respectively. The Q_H is the total hydrostatic contribution about -5.909, which can be obtained from the pressure dependence of the bandgap $\frac{dE_g^0}{dp}$ about 10.2×10^{12} **:**

$$Q_H = -\frac{C_{11} + 2C_{12}}{3}\frac{dE_g^0}{dp} \quad (5)$$

So we can obtain that the hydrostatic ΔE_H is equal to 0.36045. The shear term ΔE_s is about -0.5065 given by

$$\Delta E_s = -2b\frac{C_{11} + C_{12}}{C_{11}}e_{xx} \quad (6)$$

where b the valence-band deformation potential associated with <100> distortions is about -1.8. Hence the conduction band edge shift [2] of InAs layer is given by

$$\Delta E_C^S = \frac{2}{3}\Delta E_H = 0.2403 \text{ ev} \quad (7)$$

, the heavily-hole band edge shift [2] is obtained by

$$\Delta E_{hh}^S = -\frac{1}{2}\Delta E_s - \frac{1}{3}\Delta E_H = 0.1331 \text{ ev} \quad (8)$$

, and the light-hole band edge shift [2] is expressed by

$$\Delta E_{lh}^S = \frac{1}{4}\Delta E_s - \frac{1}{3}\Delta E_H - \frac{1}{2}\Delta(x) + \frac{1}{2}\sqrt{\frac{9}{4}\Delta E_S{}^2 + \Delta E_s\Delta(x) + \Delta^2(x)} = -0.07\text{ev} \quad (9)$$

where $\Delta(x)$ is split-off energy of InAs about 0.381 ev.

The ΔE_C^S is less than conduction band offset ΔE_C about 0.83 ev and the ΔE_{lh}^S is less than ΔE_V about 0.24 ev. So if the InAs/GaAs strained structure is constructed of InAs quantum well in Fig. 6, it can be seen that the heavy-hole band will form type-I structure and the light-hole will form type-I structure also.

Fig. 7 show cross-sectional views of the GaAs layer and InAs layer grown on GaAs substrates after wafer is etched by $NH_4OH:H_2O_2:H_2O$ etching solution in ten seconds. The cross-section image of GaAs/InAs strained layer successfully grown on GaAs substrate can be clearly observed in Fig. 7.

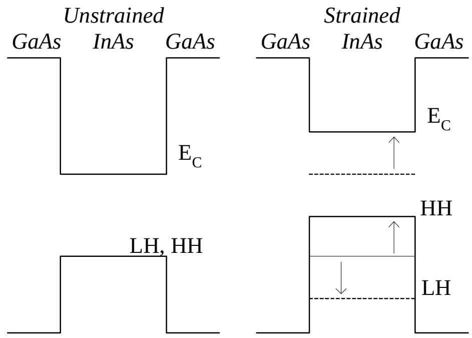


Fig. 6. The energy band of a GaAs/InAs/GaAs single quantum well

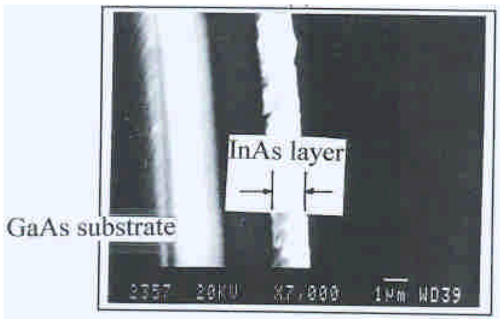


Fig. 7. The cross-section image of GaAs/InAs strained layer by SEM

6. Hall Measurement

The electrical properties of *n* and *p* type GaAs thin films with varied Si and Zn doping were respectively. The electron and hole carrier concentrations of GaAs films respectively doped Si and Zn sources were about 4×10^{17}/cm^3 and 4×10^{19}/cm^3 at the optimal V/III ratio. The *n*-like type of undoped InAs epilayer was determined by Hall measurement. A carrier concentration of 2×10^{17}/cm^3 and a Hall mobility of 2×10^3cm^2/V-sec were obtained for an undopant InAs prepared on GaAs substrate at growth temperature of 600°C at group V/III ratio of 75.

VI. SUMMARY AND CONCLUSIONS

Because of the high strain effect between InAs layer and GaAs layer, the InAs can not be grown too thick on GaAs layer. The quality of InAs layer is much better for high V/III ratio than for low V/III ratio. The quality of GaAs buffer layer is influenced by V/III ratio also. The strained effect will influence the band structure of InAs/GaAs surely. The strained effect of GaAs/InAs/GaAs hetero-structure belongs to compression strained effect. We studied the bulk characteristics of InAs/GaAs high lattice mismatch here by using metal organic chemical vapor deposition growth (MOCVD) method successfully. MOCVD can be used to precisely control the thickness and chemical composition of heteroepitaxial layers, as well as to introduce dopant species in-stiu. The thickness and interface control available in current MOCVD has produced a mature quantum well technology, and a rich array of semiconductor physics and devices for semiconductor industry in Taiwan.

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