## Monolayer Steps Formation in InP and GaInAs by OMVPE and Reduction of Resonant Energy Width in GaInAs/InP RTDs

Michihiko SUHARA, Chuma NAGAO<sup>†</sup>, Hidetaka HONJI<sup>†</sup>,

Yasuyuki MIYAMOTO<sup>†</sup>, Kazuhito FURUYA<sup>†</sup> and Riichiro.TAKEMURA<sup>†</sup> Research Center for Quantum Effect Electronics, Tokyo Institute of Technology

<sup>†</sup> Department of Electrical and Electric Engineering, Tokyo Institute of Technology O-okayama, Meguro-ku, Tokyo 152 Japan Tel:+81-3-5734-2572 Fax:+81-3-5734-2907

## Abstract

Atomically flat terraces having several hundred nanometers of the width and monolayer steps were formed on both of InP and  $Ga_{0.47}In_{0.53}As$  surfaces by OMVPE (organometallic chemical vapor deposition) for the first time. From surface observation using AFM (atomic force microscopy), growth mode were analyzed. The critical condition between step flow mode and 2D-nucleation mode in GaInAs/InP OMVPE growth was obtained. Applying the step flow mode for the growth of GaInAs/InP RTDs, remarkable reduction of resonant energy broadening to 18 meV from 51 meV was observed.

## 1. Introduction

Atomic order flatness of heterointerface is required for the realization of quantum effect devices constructed by ultrafine heterostructure. One of the attractive way to obtain the flatness is the formation of atomically flat terraces by crystal growth techniques such as organometallic vapor phase epitaxy (OMVPE) and molecular beam epitaxy (MBE). Ultimate flatten surface is obtained as wide terraces with atomic steps, whose width is intrinsically determined by misorientation angle of substrate. Such flat terraces and monolayer steps can be formed by a special growth mode called as step flow mode in contrast to 2D-nucleation mode in which islands are formed on a surface causing rough interfaces. The growth mechanism has been investigated by analyzing growth mode in MBE.<sup>1</sup>

For GaAs surfaces grown by OMVPE, the mechanism of monolayer or multilayer step formation has been studied using atomic force microscopy (AFM)<sup>2</sup>) and the mechanism of step bunching has been studied using scanning tunneling microscopy (STM)<sup>3</sup>). For GaAs, AlAs and AlGaAs OMVPE growth surfaces, the formation of wide surface terraces has been observed by AFM<sup>4</sup>). Recently atomic observation of OMVPE grown InP surface were reported<sup>5,6</sup>), however, the growth mode and the relation to heterointerface roughness have not been systematically analyzed, although InP-based materials are expected to be useful for high speed quantum devices. Previously we reported the wide terrace formation of GaInAs on InP using growth interruption in OMVPE<sup>7</sup>).

In this paper we investigated OMVPE grown InP and  $Ga_{0.47}In_{0.53}As$  surface topography using AFM. Then growth mode transition between step flow and 2Dnucleation was studied in InP on InP and GaInAs on InP surfaces, respectively, for flattening the GaInAs/InP heterointerfaces. Effect of growth mode on heterointerface roughness was estimated by measuring energy broadening of resonant level in GaInAs/InP resonant tunneling diode (RTD) fabricated by OMVPE.

## 2. Experimental

A Series of growths was performed on (100) oriented n-InP substrates whose misorientation magnitudes were within 0.2°. The pressure and flow velocity in the OMVPE reactor were 76 Torr and 3 m/s, respectively. Triethylgallium (TEG) and Trimethylindium (TMI) were used for group III source materials and AsH<sub>3</sub> and PH<sub>3</sub> were used for group V materials. Lattice mismatch of the GaInAs layer to an InP substrate was evaluated to be less than 0.05%. The flow rate of PH<sub>3</sub> and AsH<sub>3</sub> was  $4.46 \times 10^{-3}$  and  $4.46 \times 10^{-3}$  mol/min, respectively. H<sub>2</sub> was used for a carrier gas whose flow rate was on the order of 4 slm.

Before loading into the reactor, a substrate were etched in water:sulfuric acid:hydrogen peroxide (1:3:1) solution for 90 sec and rinsed in deionized water.

Ex-situ observations of a surface topography were performed by a commercial AFM (Digital Instruments Nanoscope II) in contact mode in air. More than 5 points were under the observation on a sample and maximum scanning range at each point was  $5 \,\mu$  m square.

#### 3. Grown surface observation by AFM

Prior to principal growth for AFM observation, atomic steps with 300 nm width which were formed on a substrate for each sample by performing 10 nm InP deposition with a constant condition (0.47ML/sec, 600°C) and 30 sec annealing under PH<sub>3</sub> atmosphere. On these atomically flattened substrates, InP or GaInAs were successively grown at several growth rates  $G_R$  and temperatures  $T_G$ . After 10 nm deposition, growth was interrupted and the sample was immediately cooled down to room temperature and surface topography were observed by AFM. Thermal annealing process was not performed after each growth.

Figure 1 shows typical AFM images of InP surface grown at  $G_R$  and  $T_G$ . For  $G_R$ =0.22 ML/s and  $T_G$ =600°C,



Fig.1 AFM images of OMVPE grown InP surface. (a)  $G_R$ =0.22 ML/s and  $T_G$ = 600 °C, (b)  $G_R$  =1.08 ML/s and  $T_G$ =550 °C

one-monolayer steps and terraces were observed as shown in Fig.1 (a). In this case island and/or island-like undulation at step edge were not observed. In case of  $G_R$ =1.08 ML/s,  $T_G$ =550°C, surface topography became rough and many islands and multi steps were observed as shown in Fig.1(b).

Figure 2 shows typical AFM images of  $Ga_{0.47}In_{0.53}As$  layer grown on the atomically flatten InP substrates. The condition to obtain GaInAs monolayer steps was different from these of InP. In a condition of lower growth rate such as  $G_R=0.35$  ML/s,  $T_G=650^{\circ}C$  one monolayer steps and terraces were observed as shown in Fig.2 (a). In case of low temperature and/or high growth rate such as  $G_R=0.35$  ML/s,  $T_G=600^{\circ}C$  or  $G_R=0.76$  ML/s,  $T_G=600^{\circ}C$ , islands and undulation of step edge were observed as shown in Fig.1(b).

From the results of AFM observation, a growth mode was considered. When more than one island and/or island-like nonuniform undulation at step edge were observed, the growth condition for the sample was regarded as 2D-nucleation mode. On the other hand, when uniform one monolayer steps were observed with no islands, the condition was regarded as step flow mode. So Fig1.(a) and Fig.2(a) are regarded as the growth by step flow mode and Fig.1(b) and Fig.2(b) are regarded as growth by 2D-nucleation mode.





Fig.2 AFM images of GaInAs surface. (a) R=0.35 ML/s and T=  $650^{\circ}$ C, (b)R=0.76 ML/s and T= $600^{\circ}$ C

## 4. Critical condition between step flow and 2Dnucleation mode in the OMVPE

From results of AFM observation, a critical condition for growth mode transition between step flow and 2D-nucleation in the OMVPE was analyzed based on the surface diffusion theory<sup>1</sup>). According to the theory, growth mode shifts from step flow to 2D-nucleation when the maximum supersaturation ratio on the growth surface  $\alpha_{max}$  exceeds a critical value.  $\alpha_{max}$  is derived as functions of growth rate, terrace width, partial pressure of V group gas and growth temperature and surface migration length. On the other hand the critical supersaturation ratio  $\alpha_{crit}$  for a disk-shaped nucleus is expressed as a function of surface free energy and temperature. The critical condition is given by solving the equation  $\alpha_{max} = \alpha_{crit}$ .

Figure 3 shows growth mode diagrams of InP and GaInAs as the function of G<sub>R</sub> and T<sub>G</sub>. Experimental results are also plotted. The critical condition is calculated satisfying experimental results. Here surface free energy  $\sigma$  and surface diffusion length  $\lambda_s$  of In atom are assumed as  $\sigma = 3.4 \times 10^{12} \sim 2.7 \times 10^{14}$  eV/cm<sup>-2</sup> and  $\lambda_s = 0.5 \sim 240$   $\mu$ m for P stabilized InP surface and  $\sigma = 7.8 \times 10^{14} \sim 8.4 \times 10^{14}$  eV/cm<sup>-2</sup>  $\lambda_s \sim 0.9$  nm for As stabilized GaInAs surface.

677



Fig.3 Growth mode diagram of InP and GaInAs. Plotted data  $\blacktriangle$ ,  $\triangle$ ,  $\bigcirc$ ,  $\bigcirc$  correspond to conditions shown in Fig.1(a), (b), Fig.2 (a) and (b), respectively.

# 5. Effect of growth mode on energy level broadening in GaInAs/InP RTDs

To investigate the effect of growth modes on device characteristics, GaInAs/InP resonant tunneling diodes (RTD) were fabricated by different growth modes. If any islands are formed at InP barrier layer and GaInAs quantum well layer, they causes well width variation in a lateral direction of the RTD sample. The inhomogeneous well width variations to broaden the resonant energy width<sup>11</sup>.

Fabricated GaInAs/InP RTD samples were consist of 100 nm n +-GaInAs, 260 nm n-GaInAs for an anode electrode, 2.6 nm i-GaInAs for a spacer, 8.4 nm i-InP for barrier, 4 nm i-GaInAs for quantum well, 8.4 nm i-InP for barrier, 2.6 nm i-GaInAs for a spacer, 260 nm n-GaInAs for an cathode electrode, 100 nm n +-GaInAs for top contact layer. Samples were separated by circle pattern of Cr/Au contact with the diameter of  $18 \,\mu$  m.

Two different growth modes were used for the growth of InP/GaInAs/InP quantum well structure. As RTD samples grown by step flow mode, conditions;  $T_G=650^{\circ}C$ ,  $G_R=0.22$  ML/s for InP and  $T_G=650^{\circ}C$ ,  $G_R=0.35$  ML/s for GaInAs were chosen. As RTD samples grown by 2D-nucleation mode,  $T_G=550^{\circ}C$ ,  $G_R=0.43$  ML/s for InP and  $T_G=550^{\circ}C$ ,  $G_R=0.76$  ML/s for GaInAs were chosen for 2D-nucleation mode.

To estimate resonant energy width  $(\triangle E)^{11}$ , the second derivatives  $(d^{2}I/dV^{2})$  of measured current-voltage characteristics of RTDs were calculated as shown in Figure 4. Energy-voltage conversion ratio  $(\triangle E$  vs. FWHM of  $d^{2}I/dV^{2})$  is assumed as  $0.45^{11}$ .  $\triangle E$  was evaluated as 18 meV for step flow mode and 51 meV for 2D-nucleation



Fig.4  $d^2I/dV^2$  characteristics of GaInAs/InP RTDs grown by OMVPE

mode, respectively. Remarkable reduction of  $\triangle E$  were observed by applying step flow mode. P/V ratio for both step flow and 2D-nucleation mode were around 3.

## 6. Conclusion

OMVPE grown InP and  $Ga_{0.47}In_{0.53}As$  surface topography were investigated using AFM. Atomically flat terraces and uniform monolayer steps were obtained both in InP and GaInAs growth. The critical condition between step flow mode and 2D-nucleation mode was analyzed in the OMVPE growth. Applying the step flow mode for the growth of GaInAs /InP RTDs, remarkable reduction of resonant energy broadening was observed.

## Acknowledgments

The authors thank Professor S. Arai and Associate Professors M. Asada and M.Watanabe for fruitful discussions. This work is supported by a Grant-in-Aid for Scientific Research in Priority Areas (Quantum Coherent Electronics) and by the "Quantum Nanoelectronics" Joint Research Project, both of the Ministry of Education, Science, Sports and Culture of the Japanese Government.

### References

- 1) T.Nishinaga and K.I.Cho, Jpn.J.Appl.Phys., 27, (1988), L.12-14
- 2) T.Fukui and H.Saito: Jpn.J.Appl.Phys. 29 (1990) 483
- 3) M.Kasu and N.Kobayashi, J.Cryst.Growth, 145 (1994) 120.
- M.Shinohara, M.Tanimoto, H.Yokoyama, and N.Inoue, Appl.Phys.Lett. 65, (1994) 1418.
- J.E.Epler, H.P.Schweizer, J.Pedersen and J.Sochtig, Appl.Phys.Lett. 66, (1995) 1472.
- V.Merlin, T.T.Duc, G.Younes, Y.Monteil, V.Souliere and P.Regreny, J.Appl.Phys. 78 (1995) 5048
- M.Suhara, C.Nagao, Y.Miyamoto and K.Furuya, 22nd International Symp. on compound semiconductors, FrB2-3, Cheju Island (1995)
- H.Saito, K.Uwai and N.Kobayashi, Jpn.j.Appl.Phys. 32 (1993) 4440
- 9) C.T.Foxon and B.A.Joyce, J.Crystal Growth, 44 (1978) 75
- 10)M.Kasu, H.Saito and T.Fukui, J.Crystal Growth, 115 (1991) 406
- 11)Y.C.Kang, M.Suhara, K.Furuya and R.Koizumi, Jpn.J.Appl.Phys., 34, 4417-4419 (1995)