Realization of Vertically Stacked InGaAs/GaAs Quantum Wires on V-grooves with (322) Facet Sidewalls by Chemical Beam Epitaxy

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CONTENTS

ABSTRACT

We report, for the first time, the fabrication of vertically stacked InGaAs/GaAs quantum wires (QWRs) on V-grooved substrates by chemical beam epitaxy (CBE). To fabricate the vertically stacked QWRs structure, we have grown the GaAs resharpening barrier layers on V-grooves with (100)-(322) facet configuration instead of (100)-(111) base at 450 °C. Under the conditions of low growth temperature, the growth rate of GaAs on the (322) sidewall is higher than that at the (100) bottom. Transmission electron microscopy verifies that the vertically stacked InGaAs QWRs were formed in sizes of about $200\text{Å} \times 500 \sim 600$ Å. Three distinct photoluminescence peaks related with side-quantum wells (QWLs), top-QWLs and QWRs were observed even at 200 K due to sufficient carrier and optical confinement. These results strongly suggest the existence of the quantized state in the vertically stacked InGaAs/GaAs QWRs grown by CBE.

I. INTRODUCTION

In recent years, low dimensional quantum confined semiconductor structures such as quantum wires (QWRs) and quantum dots (QDs) have attracted a great deal of attention, as they show much potential to bring forth a new class of functional devices [1]-[3]. Many growth techniques have been developed to fabricate such quantum size structures. Fine lithography, dry etching and regrowth have been used to form low dimensional structures [4],[5]. However, etch damage and contamination at the interfaces have been found to strongly reduce the quantum efficiency. In order to obtain defect-less interfaces, in-situ one-step growth processes are employed, which include the epitaxial growth on V-grooves [6],[7] or ridges [8],[9], selective area epitaxy using dielectric masks [10], or maskless growth on planar substrates [11]. Until now, a variety of studies have mostly reported GaAs/AlGaAs QWRs on the V-grooved substrates by metalorganic chemical vapor deposition (MOCVD) or molecular beam epitaxy (MBE). Multiple stacked QWRs and QDs also have advantage in applying tunneling mechanism to quantum devices. However, it is very difficult to fabricate vertically stacked InGaAs/GaAs QWRs on V-grooves because GaAs layer may grow faster in the (100) bottom plane than the (111)A sidewall plane, hence the shape of GaAs layers at the bottom changes from sharp to rounded V-grooves having wide (100) plane during growing the barrier layers. Recently, we reported on the reduction of lateral dimension of InGaAs/GaAs heterostructure on V-grooved GaAs(100) substrate by chemical beam epitaxy (CBE) [12]. By this method, we effectively control the lateral dimension of GaAs and InGaAs, respectively, using non-(111) V-grooved substrates and uncracked hydride gases.

In this paper, we report the first successful fabrication of vertically stacked InGaAs/GaAs QWRs on V-grooves by CBE. To stack the QWR vertically, the shape of the concave part of the GaAs barrier has to be maintained sharply after fabricating a QWR. We have grown GaAs resharpening barriers on V-grooves with (100)-(322) plane at growth temperature of 450°C. The growth temperature dependence of the growth-rate anisotropy of GaAs on different crystallographic orientations enable a resharpening effect, which plays a key role in fabricating stacked QWRs. The vertically stacked multiple InGaAs/GaAs QWRs were actually fabricated with a size of about $200\text{Å} \times 500 \sim 600\text{Å}$ by crystallographic selective growth and necking formation.

Using a scanning electron microscope (SEM) and transmission electron microscopy (TEM), we confirmed the resharpening effects of GaAs layer at the bottom of V-grooves and the absence of extended defects. Photoluminescence (PL) measurement has been used to verify the In composition variation resulting from the difference of In migration. Finally, temperature dependence of PL spectra clarifies the existence of the exciton ground state in the InGaAs QWRs as well as side-quantum wells (QWLs) and top-QWLs.

II. EXPERIMENT

The V-grooved substrates were fabricated on semi-insulating GaAs(100) substrates with 2° off towards (110) using standard photolithography and chemical wet etching in solution of $H_2SO_4-H_2O_2(30\%)-H_2O$ (1:8:40 in volume) at room temperature for 2.5 minutes. The fabricated V-grooves had 5 μm periods and oriented along the $[01\bar{1}]$ direction. Prior to growth, the V-grooved substrates were treated with organic solution. And then they were dipped in a concentrated HCl solution for 2 min and rinsed in deionized (DI) water, followed by etching in $H_2SO_4-H_2O_2-H_2O$ (20:1:1 in volume) for 15 seconds. From these etching processes, very smooth V-grooves closed to (322) surfaces were obtained. The substrates were then rinsed in deionized water, dried and finally loaded into the growth chamber. Fabrication process of V-grooved substrates and InGaAs/GaAs QWRs is schematically shown in Fig. 1.

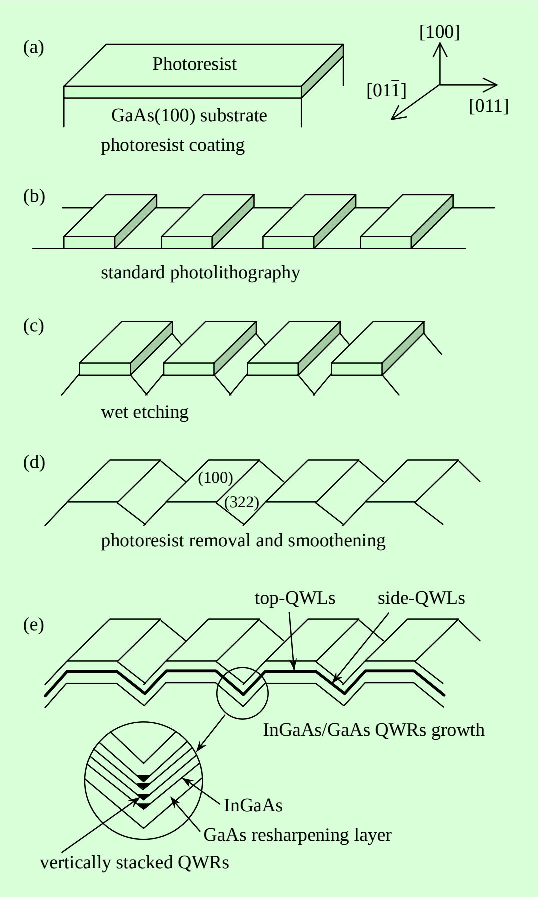

Fig. 1. Schematic fabrication sequence of InGaAs/GaAs quantum wires on the V-grooves: (a) photoresist coating, (b) patterning by standard photolithography, (c) formation of a V-grooves by wet etching in H_2SO_4-based solution, (d) photoresist removal and smoothening, and (e) stacked InGaAs quantum wires growth in CBE.

After the patterning procedure, CBE growth was performed using trimethylindium (TMIn : $(CH_3)_3In$), triethylgallium (TEGa : $(C_2H_5)_3Ga$) and uncracked monoethylarsine (MEAs : $(C_2H_5)AsH_2$) as group III and group V sources, respectively. Prior to growth, the V-grooved substrates were heated to 150°C for removal of adsorbed moisture and were annealed at

600°C for 15 min under MEAs pressure of 4×10^{-4} Torr in order to remove the surface oxide layer of the GaAs substrates. The source gases were introduced through absolute pressure controllers without carrier gas. The detailed configuration of the system used in this work has been described in a previous publication [13].

In order to investigate the resharpening effect of GaAs, we have grown GaAs epilayers on V-grooves over a wide range of growth temperatures from 400 to 600°C. The growth-rate anisotropy was examined by cross-sectional SEM and TEM. For fabricating InGaAs/GaAs QWRs, the growth temperature of GaAs and InGaAs films was kept at 450°C. The shape, thickness and defects of InGaAs QWRs were inspected by TEM. In compositions of InGaAs epilayers on each region of the V-grooves were determined by PL at 4.2 K. Temperature dependence of PL spectra was measured for optical characterization of the grown InGaAs quantum structures. For the temperature-dependent PL measurements from 77 to 200 K, the sample was mounted on a variable-temperature cryostat cooled with liquid nitrogen. The PL was dispersed by a double monochromator and measured with a liquid-nitrogen cooled Ge photodetector using a standard lock-in technique. The defocused 514.5 nm line from an Ar^+ laser with optical power density of 10∼100 mW/cm^2 was used as the excitation source.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the growth rates of GaAs on (100) top/bottom and (322) sidewall planes as a function of growth temperature. The GaAs growth rate on the (100) planes increased with increasing growth temperature. On the other hand, the growth rate on sidewalls decreased with increasing growth temperature from 450 to 600°C. Otherwise, at lower temperature of 400°C, we could not grow GaAs epilayer. The anisotropy factor defined by the ratio of the growth rate on the sidewall to the (100) top plane is plotted against growth temperature in Fig. 2(b). The anisotropy factor decreased monotonically with increasing growth temperature. This factor reaches unity at the temperature of 450°C. This means that, if a GaAs is growth on a V-grooved substrate at growth temperature above 450°C, the V-shaped geometry is lost and becomes round at the bottom, whereas the shape of the bottom V-grooves remains very sharp at 450°C. This result suggests that the lateral migration length of Ga atoms from the (322) sidewalls to the (100) bottom decreases with decreasing growth temperature. The (322) plane combines with step-edge atom with one dangling bond per a atom and terrace atom with two dangling bonds per a atom. In contrast the (111)A plane has a dangling bond per a atom. This means that the density of dangling bonds of (322) plane has greater than (111)A plane. Due to the use of (322) plane having a greater number of arsenic dangling bonds than (111)A plane, the growth rate of GaAs on (322) sidewalls is effectively enhanced by the suppression of Ga migration. These effects give a possibility of using a GaAs layer as resharpening layer in vertically stacked multiple InGaAs/GaAs QWRs. If the second GaAs barrier grows thick enough, the shape of V-groove at the concave part will become sharp, resulting in fabricating multiple stacked InGaAs/GaAs QWRs.

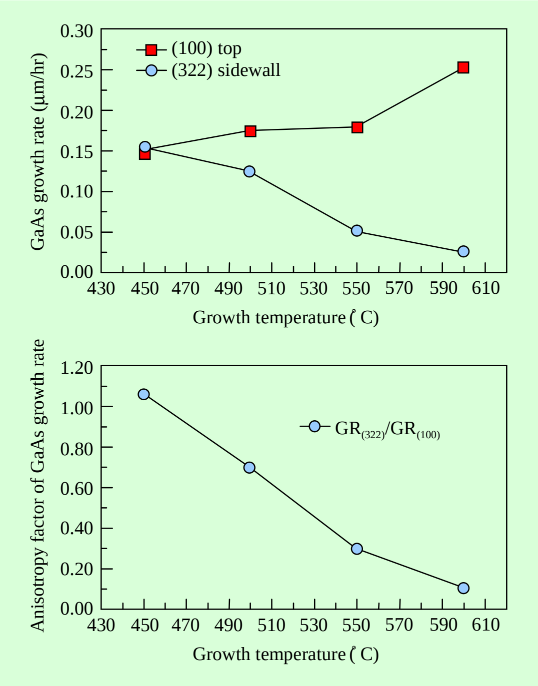

Fig. 2. Growth temperature dependence of (a) growth rate, and (b) anisotropy factor of GaAs on the (100) bottom and (322) sidewall. Anisotropy factor of GaAs growth rate monotonically decreases with increasing growth temperature.

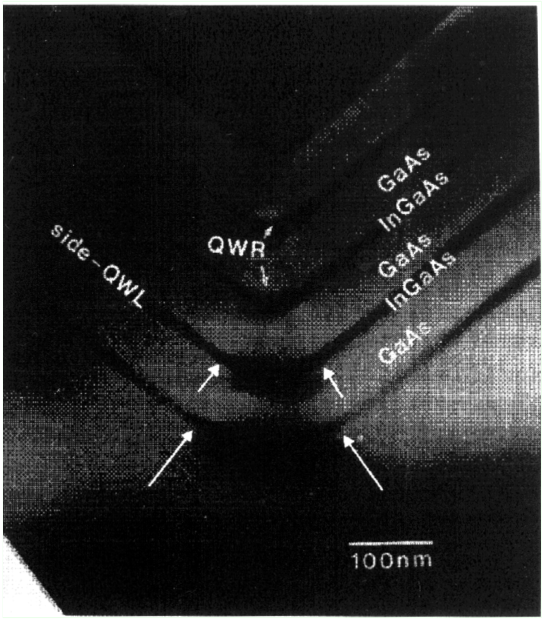

Fig. 3. Cross-section transmission electron microscopy image showing resharpening effect of GaAs on V-grooves (lower two images) and vertically stacked InGaAs/GaAs quantum wires (upper two images). Dark areas show InGaAs epilayers.

Figure 3 shows a typical cross-sectional TEM image of InGaAs/GaAs QWRs grown on a V-groove patterned GaAs substrate. SEM photographs reveal a cross-sectional view of about 2.5 μm-wide and 1.6 μm-deep V-grooves of InGaAs/GaAs which were aligned along the $[01\overline{1}]$ direction with a 4.5 μm period. Even though the top-QWLs are not seen in the TEM image of Fig. 3, there are three quantum structures of bottom QWRs, top-QWLs and side-QWLs. The InGaAs/GaAs QWRs were fabricated in-situ by CBE at a growth temperature of 450°C. This fabrication technique is essentially based on the crystallographic selective growth and migration of adspecies and was reported in our previous result [12]. At 450°C, the migration of Ga adatoms is suppressed while that of In adatoms is enhanced, resulting in an efficient confinement of InGaAs layers. As shown in Fig. 3, the (100) bottom plane is disappearing and the V-groove becomes sharp because the growth rate of GaAs on the bottom is less than that at the sidewall. In case of the V-groove system, in contrast to ridge or planar system, the plane with lower growth rate is disappeared. On the other hand, the InGaAs growth rate at the bottom is much higher than that on the sidewall. As we discussed earlier, to sharpen the bottom of the V-grooves, it is necessary that adatoms incorporate into the sidewall surface much more easily than into the bottom surface so that the growth rate of sidewall is higher than that at the bottom. As clearly shown in the lower two shapes of Fig. 3, the GaAs barrier layers have the function of resharpening the shape of the (100) bottom of the V-grooves. This result also utilized the mechamism of relatvie migration length of group III (In>Ga>Al) as with (Al,Ga)As resharpening layer of crescent-shaped GaAs/AlGaAs multiple QWRs [7],[14], where the GaAs layer shows a fast planarization of the V-groove by creating a new (100) surface at the bottom due to the fast surface migration of Ga atoms. In contrast, in this paper, we achieved GaAs barrier layers which sharpened to the shape of V-grooves by restraining the migration of Ga atoms with the use of low growth temperature of 450°C and (322) sidewall planes. For the growth of active layers, it is important that the QWRs separate from side-QWLs. In order to confine the lateral dimension effectively, many studies have utilized, in variety of ways, the difference in the growth rate on various crystal facets [15]-[17]. We have also achieved a very large anisotropy of growth rates on different crystal planes (sidewall and bottom) employing the non-(111) V-grooved substrates and necking areas (arrows in Fig. 3) [11]. The upper two shapes of Fig. 3 distinctly show the crescent-shaped InGaAs QWRs with a size of about 200 Å × 500 ~ 600 Å at the bottom formed by growth anisotropy and necks. The maximum thickness of QWRs is about three times that of the 70Å-thick QWLs on the adjacent sidewalls. This shape of InGaAs QWRs, due to the migration of group III atoms from the sidewall to the bottom, is very similar to that of GaAs QWRs by MOCVD. However, we report, for the first time, the multiple InGaAs QWRs vertically stacked on V-grooves using GaAs as resharpening layers. The TEM photograph also indicates that there are no defects, such as dislocations, stacking faults and twins, neither at the bottom of V-grooves nor on the sidewalls of the grown sample. The defect-free character is important for optical and electrical properties of devices made with such QWRs. Furthermore, in comparison with MOCVD methods which have been widely used to fabricate QWRs on V-grooved substrates, the CBE growth technique has the advantage of thickness control, and hence the fabrication of fine quantum structures can be expected.

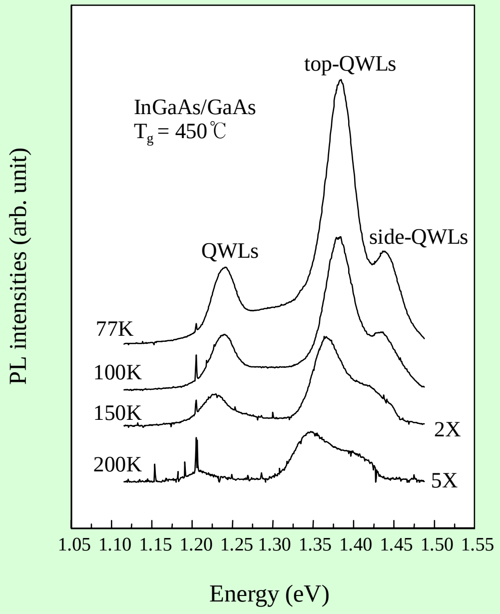

Fig. 4. Temperature dependent photoluminescence spectra of the sample including InGaAs/GaAs quantum wires. Two peaks at high energy side originate from the side-quantum wells (QLWs) and top-QLWs as the third peak having the lowest energy comes from the bottom quatum wires. Three distinct peaks are still observed even at 200 K.

Figure 4 presents temperature-dependent PL spectra of the InGaAs/GaAs QWRs illustrated in Fig. 3. We observe three distinct peaks related with side-QWLs, top-QWLs and QWRs. The spatial origin of the luminescence signals is verified in relations with film thickness, compositions, peak positions and intensities. The thickness of side-QWLs is thinner than that of top-QWLs or bottom QWRs owing to surface migration of InGaAs adatoms on the sidewall to the bottom- and top-layers. Examination of the PL spectra at 77 K indicates that the highest-energy luminescence at 1.439 eV originates from the InGaAs side-QWLs with a thickness of about 70 Å. Asymmetry found in thickness of side-QWLs might origin from the use of 2° tilted substrate. The PL peak with strong intensity at 1.384 eV comes from the top-QWLs. The strong PL peak of top-QWLs is obtained over a large portion of the entire sample area, and it suggest that the layer is nearly free of defects which could have been induced from chemical wet etching processes. The PL peak with the lowest energy can be allocated to be QWRs at the bottom. The width of QWRs (upper two images in Fig. 3) is about 500∼ 600 Å which is wide to the extent that the effect of lateral confinement is weak. Therefore the peak at the lowest energy side with broad FWHM of 30 meV included both QWRs' peaks and bottom QWLs' peaks (lower two images in Fig. 3). The peak energy, 1.240 eV, is far lower and stronger than what we expected. We consider that the lower energy is attributed to two reasons. First, as mentioned above, the vertical thickness of QWRs at the bottom increases due to surface migration of group III species from (322) sidewalls to the bottom of V-grooves. This result is consistent with the red shift of QWRs PL peak and side-QWLs PL peak at the highest energy. Second, the In composition at the bottom region significantly increases because the surface migration length of In is larger than that of Ga [18],[19]. The InGaAs layers in every bottom intersection are darker than those on the sidewalls, showing strong contrast, as demonstrated by Fig. 3. This suggests that the In composition of InGaAs at the bottom is higher. Although the full width at half maximums of these luminescence peaks have somewhat larger values of about 20 to 30 meV due to the fluctuation of well thickness, these three peaks, including the QWRs peak are clearly observed even at a temperature of 200 K. In general, the optical properties of strained epilayers over a critical thickness are degraded due to misfit dislocations. Therefore, it is difficult to measure the luminescence of InGaAs QWRs at the bottom of V-grooves due to the large strain of about 2%. However, we observed a very strong and separated luminescence peak at 1.240 eV, which comes from the InGaAs QWRs in spite of its thickness of about 200 Åand its small portion of the entire area, as shown in Fig. 3. This phenomenon indicates a strong optical confinement effect in the defect-free InGaAs QWRs. The crescent shape and necks also give reasonable subband separations. These results suggest that optically high-quality stacked InGaAs QWRs were successfully grown on the V-grooves using GaAs resharpening barrier layers by CBE.

IV. CONCLUSION

We report the resharpening effect of GaAs layers grown on V-grooved substrate by CBE. We achieved GaAs resharpening layers in such a way that low growth temperature and (322) sidewall efficiently enhance the growth rate of GaAs on the sidewall owing to the suppression of Ga migration. Under these growth conditions, we successfully fabricated vertically stacked InGaAs/GaAs QWRs by CBE. TEM and PL measurements show that multiple crescent-shaped InGaAs/GaAs QWRs with a size of 200 Å × 500 ~ 600 Å were formed at the bottom of V-grooves. Three distinct PL peaks related with side-QWLs, top-QWLs and QWRs could be identified even at temperature of a 200 K. Furthermore, in spite of large thickness and small area, QWRs show very strong PL intensity, resulting from sufficient carrier and optical confinement in InGaAs QWRs.

ACKNOWLEDGMENTS

We would like to thank Prof. S. J. Park and T. Y. Soeng in KJIST for fruitful discussion and TEM measurements. We also wish to express our thanks to Dr. K. S. Lee for his support in the PL analysis. This work was supported in part by the Ministry of Information and Communications, Korea.

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