

Growth and Characterization of AlGaAs Nanowires on Insulating Al₂O₃ Layers by Selective-Area Metal-Organic Vapor Phase Epitaxy

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1. Introduction

Semiconductor nanowires (NWs), which are mostly fabricated by vapor-liquid-solid (VLS) method using metal nano-particles as a catalyst, have attracted much attention in the research field of nanoscale electronic and photonic devices. We have fabricated III-V compound semiconductor NWs by using selective-area metal-organic vapor phase epitaxy (SA-MOVPE), and realized GaAs/AlGaAs core-shell NWs and AlGaAs nanotubes [1]. Fabrication of semiconductor NWs, in addition, has recently been demonstrated on insulating layers, e.g. GaN nano-rods on amorphous glass substrates [2] and Ge NWs on SiO₂ layers on Si substrates by the VLS method [3], because they are promising for the fabrication of light-emitting diodes (LEDs) on glass substrates. However, these NWs grown by the VLS method on insulating layers have much poor size uniformity, and are distributed randomly on the substrates. In this paper, we present the growth and characterization of semiconductor NWs on insulating Al₂O₃ layers by SA-MOVPE. It has been reported that Al₂O₃ thin layers deposited by atomic layer deposition (ALD) are crystallized after the annealing process [4]. In addition, research on epitaxial growth of single-crystalline γ -Al₂O₃ layers has been reported [5, 6]. In order to realize semiconductor NWs well-controlled on insulating substrates, therefore, we demonstrate and investigate epitaxial crystallization of Al₂O₃ layers deposited by ALD on Si (111) substrates by thermal treatment and SA-MOVPE of AlGaAs NWs on the Al₂O₃ layers.

2. Experimental Procedures

The fabrication processes for AlGaAs NWs on Al₂O₃ layers are summarized in Fig. 1. After the deposition of Al₂O₃ layers by ALD (Fig. 1(a)) and 20-nm-thick SiO₂ mask by plasma-enhanced chemical vapor deposition, or PE-CVD, (Fig. 1(b)), the initial SiO₂ mask openings, whose diameters were 50 to 200 nm, were formed using electron beam lithography (EBL) and reactive ion etching (RIE), as shown in Fig. 1(c). The thicknesses of the Al₂O₃

layers were changed from 5 to 10 nm on Si (111) substrates. Before the growth of the NWs, Si (111) substrates with Al₂O₃ layers on top were annealed in hydrogen, H₂, atmosphere in our MOVPE reactor. The annealing temperatures and time for the experiments of the crystallization of Al₂O₃ layers were changed from 875 to 925 °C and 5 to 15 min, respectively (Fig. 1(d)). Finally, AlGaAs NWs were grown by SA-MOVPE on the Al₂O₃ layer surfaces in the initial mask openings of the SiO₂ layers (Fig. 1(e)). (CH₃)₃Ga (TMGa), (CH₃)₃Al (TMAI) and AsH₃ diluted in H₂ were used for source materials. The partial pressures of TMGa, TMAI, and AsH₃ were [TMGa] = 1.8×10^{-6} , [TMAI] = 3.2×10^{-7} , [AsH₃] = 1.2×10^{-3} atm, respectively. Scanning electron microscopy (SEM) was used for the observations of AlGaAs NWs and surface morphology, and transmission electron microscopy (TEM) was used for the cross-sectional lattice images of the interfaces in detail.

3. Results and Discussion

Figure 2 shows a typical SEM image of AlGaAs NWs on the Al₂O₃ layers in the initial mask openings. The annealing in H₂ atmosphere was carried out for this sample at the temperature of 875 °C for 15 min. The average height and diameter of the NWs were estimated to be about 350 and 100 nm, respectively. Figure 3 is a summary of the cross-sectional TEM observation results for the sample of the AlGaAs NWs shown in the SEM image of Fig. 2. We observed the cross-sectional images nearly in the middle of the NWs from the <0-11> direction, as schematically illustrated in Fig. 3(a). As shown in Fig 3(b), we observed the thin layers with a bright contrast between AlGaAs NWs and Si (111) substrates in the initial SiO₂ mask openings. The thin layers are supposed to be insulating Al₂O₃ layers deposited by ALD, and the thickness of the layers was typically estimated to be 3 to 4 monolayers (MLs). We observed that the 3-ML-thick Al₂O₃ layers under the AlGaAs NWs were possibly single crystalline, as shown in Fig. 3(c). On the other hand, we confirmed that the

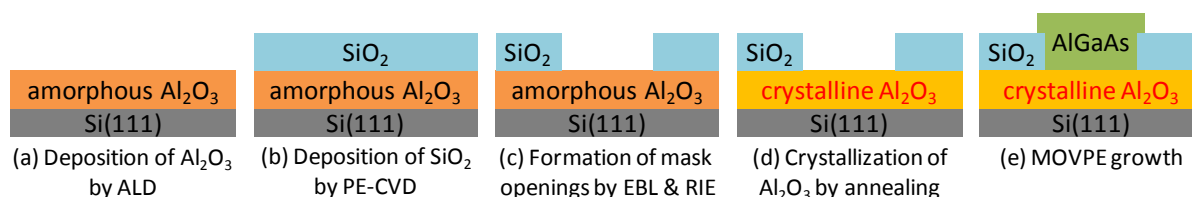


Fig. 1 Schematic illustrations of NW fabrication and Al₂O₃ crystallization process

Al_2O_3 layers between the SiO_2 mask layers and the Si (111) substrates were amorphous, and the thickness was about 4.5 nm, which was nearly the same as the designed thickness of Al_2O_3 layers, i.e. 5 nm, by ALD. Therefore, we concluded that approximately 4 nm in thickness of the Al_2O_3 layers was etched off in the initial SiO_2 mask openings owing to poor controllability of etching rate in the RIE process in the current work. AlGaAs NWs have been grown on the single-crystallized Al_2O_3 layers, but there is still a possibility that the NWs are formed partly from the small areas of the unintentionally-exposed Si (111) surfaces possibly caused by poor uniformity of the etching rates within the initial SiO_2 mask openings.

Next, we changed the designed thickness of the Al_2O_3 layers from 5 to 10 nm. Figure 4 shows the SEM images for the typical results of the SA-MOVPE growth AlGaAs on the Al_2O_3 layers with the designed thickness of 10 nm. The annealing time in H_2 atmosphere for these samples was set to 5 min. After annealing at 875 °C, some of the hexagonal AlGaAs NWs were grown partly within the initial SiO_2 mask openings. When we increased the temperature from 875 to 925 °C, hexagonal or tetrahedral AlGaAs nanostructures were formed, and the initial mask openings were completely filled with the AlGaAs nanostructures. Evaluating the actual thickness of the Al_2O_3 layers in the initial SiO_2 mask openings from the cross-sectional observations in Fig. 3, we roughly estimated the thickness of the Al_2O_3 layers to be approximately 5 to 6 nm for the samples in Fig. 4. Therefore, no exposure of the Si (111) surfaces were possibly expected for the samples in Fig. 4. In the case of the annealing at 875 °C in Fig. 4 (a), most of the Al_2O_3 layers might be still amorphous, but some parts of the Al_2O_3 layers within the initial SiO_2 mask openings were crystallized. On the other hand, in the case of the annealing at 925 °C in Fig. 4(b), most of the Al_2O_3 layers might be crystallized within the initial SiO_2 mask openings, since the AlGaAs nanostructures were formed in all the initial SiO_2 mask openings.

4. Summary

We demonstrated and investigated epitaxial crystallization of insulating Al_2O_3 layers deposited by ALD on Si (111) substrates by thermal treatment and SA-MOVPE of AlGaAs NWs on the Al_2O_3 layers. We found that the growth of nanostructures on Al_2O_3 layers strongly depended on the thickness and the annealing temperature of Al_2O_3 layers.

References

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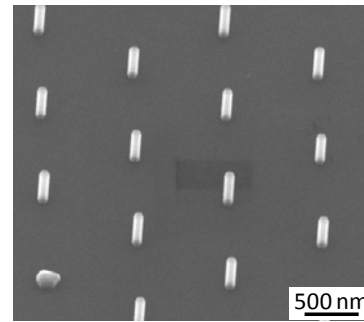


Fig. 2 45° tilted SEM image of AlGaAs NWs on 5-nm-thick Al_2O_3 layers.

(a) Top

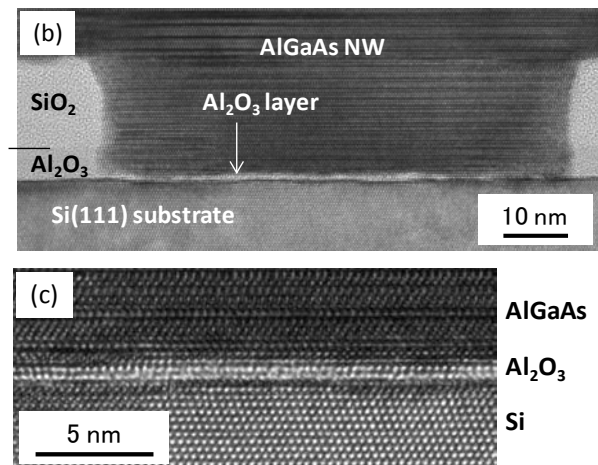
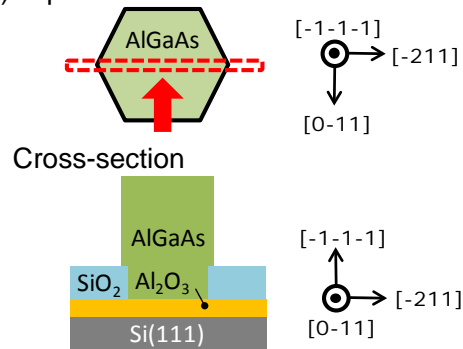


Fig. 3 (a) Schematic illustrations for cross-sectional TEM observations of NW. (b) Cross-sectional TEM image and (c) lattice image around the interface between AlGaAs NW and Al_2O_3 layers.

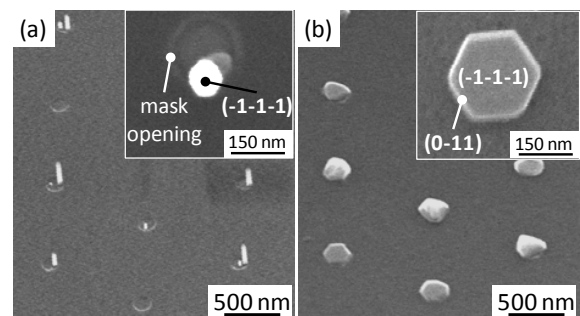


Fig. 4 45° tilted SEM images of AlGaAs nanostructures on 10-nm-thick Al_2O_3 layers annealed at (a) 875 °C, (b) 925 °C.