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Selective, Maskless Growth of InSb on a Selenium-Treated GaAs by Molecular Beam Epitaxy

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InSb nanoscale crystal islands were grown on a Se-terminated GaAs substrate by molecular beam epitaxy (MBE). In situ synchrotron radiation photoelectron spectroscopy studies for InSb island formation on this surface show that Sb atoms do not chemisorb directly on the Se-terminated GaAs surface whereas, in the sequential deposition of In and Sb, InSb islands are formed. Furthermore, it is found that the InSb islands with an average size of 30 nm and with a density of the order of 10^{10} cm^{-2} can be obtained.

1. INTRODUCTION

Growth of semiconductor nanoscale crystals on semiconducting substrates has the attractive possibility of realizing the quasi-zero dimensional quantum well structures. There are only a few reports on fabricating this type of nanoscale crystals without using photolithography, dry etching and regrowth. These are to grow fractional monolayers on tilted substrates, to grow a lattice-mismatched epilayer in hydride vapor phase epitaxy, or to utilize the droplet formation in molecular beam epitaxy (MBE).[1-5] Chikyow et al reported microcrystal growth of GaAs on ZnSe and Se-terminated GaAlAs surfaces.[4,5] However, the nucleation mechanism responsible for this growth and the chemical bondings at the interface between the microcrystal and the substrate are not clearly elucidated.

In this work, we have employed synchrotron radiation photoelectron spectroscopy (SRPES) to clarify the chemical bonding evolution upon the nanoscale island growth of InSb on Se-passivated GaAs surfaces, which is responsible for the nano-structure formation. These InSb-grown nano-structures have also been characterized by high-resolution scanning electron microscopy (HRSEM) and atomic force microscopy (AFM).

2. EXPERIMENTAL

The samples used here were n-type GaAs(001) wafers (Si doped) with a carrier density of 1×10^{18} cm^{-3}. The GaAs wafers etched by dipping in a commercial alkaline based etchant were attached to a Mo sample holder with In solder and then placed in a vacuum chamber connected to both a surface analysis and MBE chamber. The GaAs was then heated in an As overpressure for about 10 min at 600°C to desorb any remaining oxides. The temperature was then lowered to about 550°C where a 100-nm thick GaAs epitaxial layer with an As_4/Ga flux ratio of about 10 was grown and a fine streaky 2×4 reflection high energy electron diffraction (RHEED) pattern was observed. In the Se treatment, Se beam flux was supplied to the As-stabilized GaAs surface at 470°C for 5 minutes where the surface structure changed to 2x1 RHEED pattern. This reconstructed structure implies a Se-passivated GaAs surface.[6] In the InSb growth, conventional effusion cells containing elemental In and Sb were used as sources.

SRPES measurements were performed in situ in the surface analysis chamber connected to the MBE system, located at the Photon Factory on beamline BL-1A in Tsukuba. The photon energy was adjusted to 90.0 eV using a grating/crystal monochromator calibrated by directly measuring the Au Fermi edge. The advantages of synchrotron radiation over conventional X-ray photoelectron spectroscopy in the analysis of Se is that the Se3d cross section increases by over a factor of 50 as the incident photon energy is changed from 1486.6 eV (Al Kα) to 90 eV, and that the electron mean free path decreases from about 1.5 to 0.5 nm. The grown nanoscale island structures were characterized by HRSEM and AFM observations.

3. RESULTS AND DISCUSSION

In order to investigate the dependence of Sb adsorption on the topmost surface atoms of the GaAs substrates, two kinds of samples

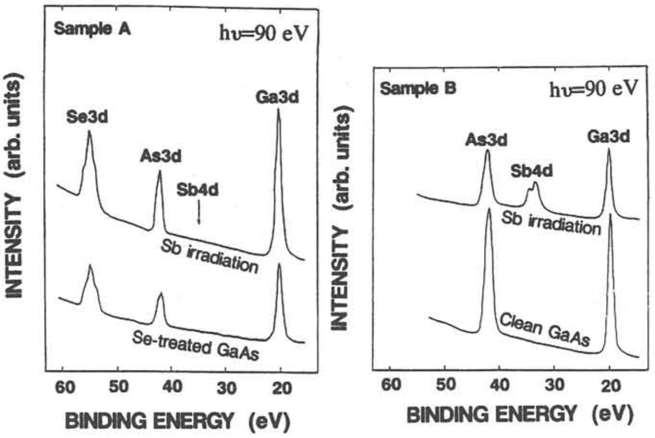

Fig. 1 Core-level photoelectron spectra before and after Sb deposition at 400^{o}C (a) with Se-treatment and (b) without Se-treatment.

were prepared: 1) the Se-passivated GaAs surface obtained by the Se treatment as mentioned above (Sample A) and 2) the As-stabilized GaAs surface only after growing a GaAs buffer layer (Sample B). After Sb_4 beam flux irradiation on both two kinds of samples at 400^{o}C for 20 seconds, the core-level photoelectron spectra were measured. As shown in Fig. 1(a), on the Se-passivated surface the Sb4d peak at around 32 eV of binding energy did not appear at all after Sb_4 irradiation, whereas on the As-stabilized surface the Sb4d peak appeared, suggesting that Sb atoms do not chemisorb on the Se-passivated GaAs surface whereas Sb atoms are bonding with surface atoms on the GaAs surface. Recently, we proposed a Ga-vacancy Ga_2Se_3 structure model for the Se-passivated GaAs(001) as schematically shown in Fig. 2.[7] Thus, it is thought that the topmost Se atoms bonding to Ga atoms do not react with impinging Sb atoms. This result is consistent with the thermodynamical data, which indicate that the heat formation of GaSb is less than that of GaSe.[8] In contrast, Harrison et al. calculated a substitution energy of Sb in the arsenic site of GaAs of $E_{GaAs}(Sb_{As})$=2.17 eV, suggesting that Sb atoms do not chemisorb on the GaAs surface,[9] However, Sb was deposited on the GaAs surface as shown in Fig 1(b). This result can be explained qualitatively by considering the high desorption rate of As species from the substrate surface at this temperature.[10]

Table I Relative peak intensities

	As3d	Ga3d
Before In depo.	1 *fixed*	1 *fixed*
After In depo.	0.80	0.82
After Sb depo.	1.02	1.07

From the mentioned-above result, when indium droplets are formed on the Se-terminated GaAs surface beforehand, InSb microcrystals are expected to be selectively grown by In incorporating Sb adatoms under Sb_4 beam flux irradiation at an appropriate substrate temperature. Core-level photoelectron spectra were measured before and after In deposition on the Se-terminated GaAs surface at 400^{o}C and after subsequent Sb deposition at the same temperature. The As3d and Ga3d spectral feature changes can not be observed through the In and Sb depositions, indicating that In

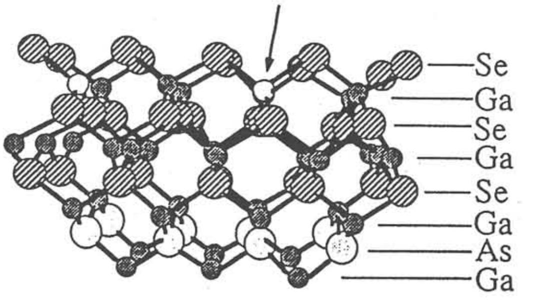

Fig. 2 Ga-vacancy Ga_2Se_3 structure model for the Se-passivated GaAs(001)

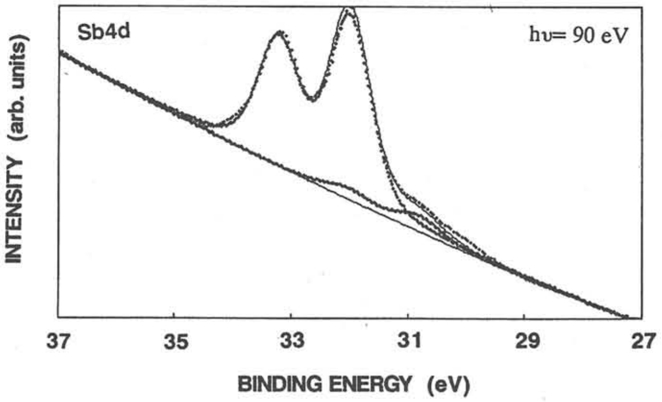

Fig. 3 Sb4d photoelectron spectrum after In and Sb deposition on the Se-terminated GaAs surface at 400^{o}C.

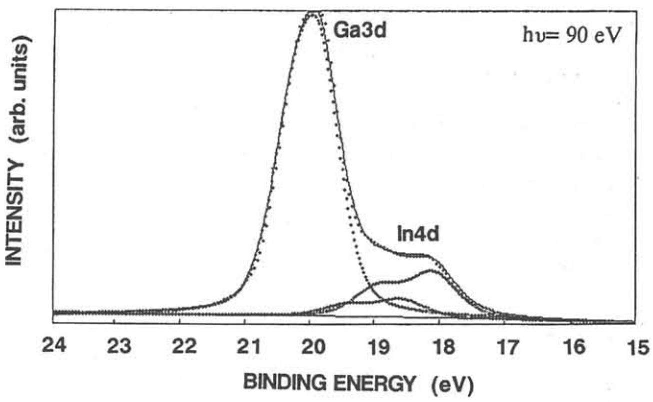

Fig. 4 Ga3d and In4d photoelectron spectrum after In and Sb deposition on the Se-terminated GaAs surface at 400^{o}C.

and Sb do not react with GaAs. Indeed, as shown in Fig. 3, the Sb4d spectrum after In and Sb deposition can be resolved into the two components comprising the main In-Sb bonding states (higher energy peak) and the surface states (lower energy peak). Figure 4 shows the Ga3d and In4d spectrum after In and Sb deposition. From peak fitting, the Ga3d peak and the two In4d peaks attributed to In-Sb bonding ($In4d_{5/2}$ at 18.1 eV) and to probably In-Se bonding ($In4d_{5/2}$ at 18.6 eV), respectively, are clearly resolved. Furthermore, as shown in Table I, the intensities of both the Ga3d and As3d peaks decrease with In deposition and then recover up to the initial values. From these results, one possible mechanism to explain the InSb formation behavior is as follows. The In overlayer is grown in the nearly laminar mode, and then Sb atoms diffuse across the Se-terminated surface until bonding to In or desorption. Consequently InSb islands are formed due to the highly lattice mismatch (14.6%).

Figure 5 shows an HRSEM image for the sample after In and Sb deposition at 400°C. Many rectangular shaped crystals are observed on the Se-terminated GaAs surface. It is found that these crystals expand to [-110] direction compared with [110] direction. This result could be understood qualitatively as that, in the case of MBE growth on GaAs(001), the lateral growth rate along [-110] is larger than that along [110] due to anisotropic surface diffusion length of Ga during MBE growth. Figure 6 shows an AFM image for the sample after growing InSb on the Se-terminated GaAs surface at 200°C. In this case the anisotropic feature, as observed in Fig. 5, has completely disappeared, which may be caused by the temperature dependence of the strength of diffusion length anisotropy. From this result, the InSb islands with an average size of 30 nm and with a density of the order of 10^{10} cm^{-2} were obtained.

In conclusion, InSb nanoscale crystal islands were grown on the Se-terminated GaAs substrate by MBE. The chemical bonding evolution upon the InSb island growth was clarified by the in situ SRPES studies. Sb atoms do not chemisorb directly on the Se-terminated GaAs surface whereas, in the sequential deposition of In and Sb, InSb islands are formed. Furthermore, it is found that the InSb islands with an average size of 30 nm and with a density of the order of 10^{10} cm^{-2} can be obtained.

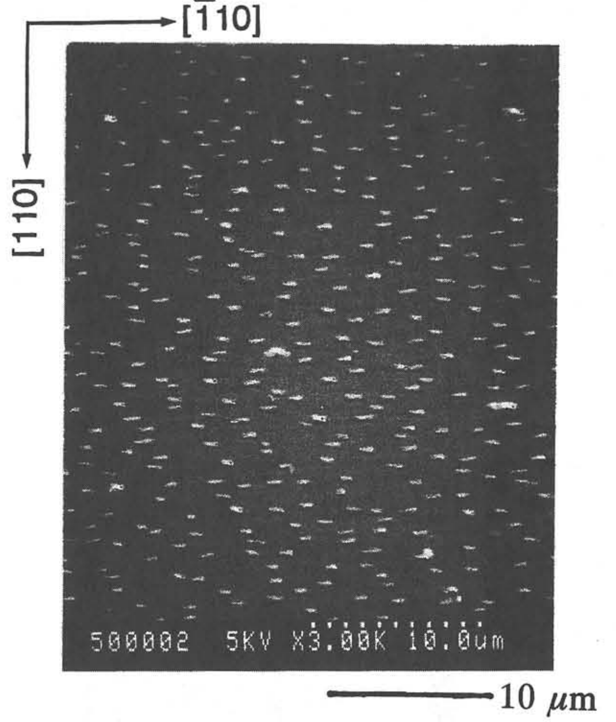

Fig. 5 HRSEM image for the sample after In and Sb deposition at 400°C.

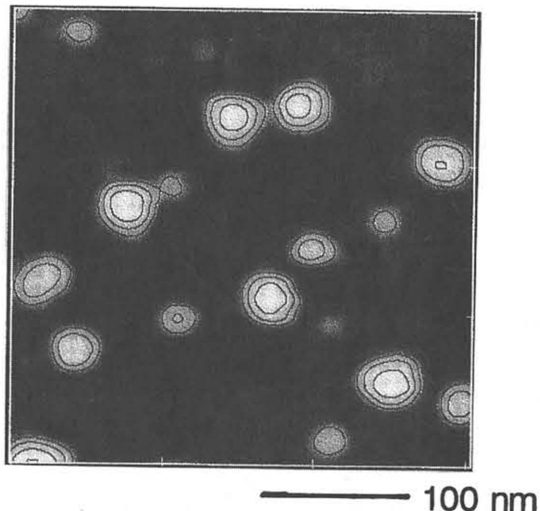

Fig. 6 AFM image for the sample after In and Sb deposition at 200°C. The contour interval is 2 nm.

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