X-ray CTR scattering measurement to investigate the formation process of InP/GaInAs interface

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Abstract. It has been successfully shown that X-ray CTR scattering measurement is a good tool to study the formation process of the InP/GaInAs interfaces grown by OMVPE even when the measurement is conducted after the growth. Using the X-ray CTR scattering measurement, the contributions of effects, i.e., surface roughness, adsorption of atoms, and exchange of atoms, on the formation process of InP/GaInAs interface were able to be discussed. The adsorption of As atoms on the GaInAs surface was shown to have the largest contribution on the abruptness of the interface.

1. Introduction

When hetero-structures of III-V compound semiconductors are grown by OMVPE (organometallic vapor phase epitaxy), which is one of the most advanced techniques to grow semiconductor crystals, a diffusive interface is generally observed, though it is designed to be abrupt at an atomic scale. For example, in InP/GaInAs hetero-structures grown by OMVPE, it is well known that As atoms distribute in InP layers grow on GaInAs layers. The distribution is often a severe problem for the layer structures in the most advanced devices, of which thickness and abruptness of the interfaces should be controlled in ML(molecular layer) levels. Therefore the mechanisms which cause the distributions at the interfaces should be investigated to find ways to control the distributions.

In order to investigate the distributions of atoms at the interfaces, we have used X-ray crystal truncation rod (CTR) scattering measurement[1-3] and demonstrated that the X-ray CTR scattering measurement is a very powerful technique to analyze the interface structures in hetero-epitaxially grown III-V compound semiconductors[4-7]. The X-ray CTR scattering measurement can reveal layer thicknesses, composition profiles, surface roughness, and lattice constants at an atomic scale. Using the X-ray CTR scattering measurements, we have shown that not only the distributions of group-V atoms but also group-III atoms are quite different from the designed ones.

In this work, we analyze the InP/GaInAs hetero-interfaces grown by OMVPE using the X-ray CTR scattering measurement, and discuss the process occurred at growth front of the forming interface from the results of the ex-situ X-ray CTR scattering measurement.
2. Experimental

The InP/GaInAs/InP structure samples were grown using a vertical reactor OMVPE on n-type InP (001) substrates. The pressure in the reactor was kept at 76.0 Torr during the growth. Typical sample structure is schematically drawn in Fig. 1. TEGa (triethylgallium), TMIn (trimethlindium), TBAs (tertiarybutylarsine) and TBP (tertiarybutylephosphine) were used as precursors and hydrogen was used as a carrier gas. Total flow rate of the hydrogen was kept at 4000sccm. The V/III ratio was 20 and the growth temperature was 620°C. The Ga composition of the GaInAs layer was designed to be 0.47.

In order to study the formation process of the InP/GaInAs interfaces, several sets of samples were prepared by changing the growth rate of the InP and GaInAs layers and the source-gas-exchange sequence at the InP/GaInAs interface. Figure 2 shows examples of measured and theoretically calculated X-ray CTR scattering spectra of a set of samples prepared by changing the growth interruption time in TBAs atmosphere.

The X-ray CTR scattering measurement was performed at the BL6A of the Photon Factory in the High Energy Accelerator Research Organization at Tsukuba. Wavelength of the X-ray was set at 1.0 Å by a Si(111) monochromator. A Weissenberg camera was used to record the X-ray CTR scattering intensity around 002 Bragg diffraction spot of InP utilizing CCD camera as a detector.

3. How to analyze the formation process of the interfaces

The analysis of the measured X-ray CTR scattering spectra gives distributions of atoms at the interfaces as shown in Fig. 3. Comparing the obtained distributions of atoms from the samples prepared with different growth conditions, it should be discussed how the distribution was caused. In other words, the formation process of the interfaces should be discussed with the results of the X-ray CTR scattering measurement after the growth.

In this work, the origin of the distributions of As and Ga at the interfaces was analyzed with the assumptions that major mechanisms which cause the distributions in InP layers on GaInAs were 1) roughness of the GaInAs surface, 2) adsorption of As and Ga atoms on the GaInAs surface, and 3) exchange of atoms between the grown GaInAs surface and the growing InP layers.

From the distributions of atoms in the samples prepared with different source-gas-exchange sequences, the adsorption and desorption rates of As were successfully analyzed with rate equation as

\[
\frac{dC}{dt} = f - \frac{C}{\tau}
\]

Fig. 1. Schematically drawn sample structure. All the samples were grown by OMVPE.

Fig. 2. Examples of measured and theoretically calculated X-ray CTR scattering spectra of a set of samples prepared by changing the growth interruption time in TBAs atmosphere. Black dots show measured data, and gray lines show theoretically calculated CTR spectra. InP 002 Bragg peaks should be at l=2.0 although they were truncated in the figure since the peaks were too high.
where $C$ was concentration of atom adsorbing on a surface, $f$ was adsorption velocity, and $\tau$ is lifetime of As on the surface. Curves obtained by solving the rate equation were fitted to the measured dependence of the amount distributing As on growth interruption time. The results showed that the GaInAs surface was covered by 4ML of As just after the growth. The lifetime, $\tau$, was analyzed to be 23 and 22s in TBAs and H$_2$ atmosphere, respectively.

On the other hand, the distribution of Ga showed almost no change with the change of the source-gas-exchange sequence. The amount of Ga distributing in InP layer was always about 0.5ML. The result suggested that almost no except Ga atoms adsorbed on the GaInAs surface and the 0.5ML of Ga distribution was caused by the exchange of atoms and/or the roughness of the GaInAs layer.

Contribution of the exchange of atoms on the formation process was able to be investigated by analyzing the distributions of atoms in the samples prepared with different growth rates. On the analysis, it was assumed that the exchange of atoms occurs only in two molecular layers, i.e., the surface one molecular layer and the second molecular layer just under the surface layer. It was also assumed that two exchange processes, i.e., from the second layer to the surface and from the surface to the second layer, could be asymmetric. The results showed that the contribution of the exchange process was very small. It was only about 0.1ML in the total amount of As and Ga distributing in InP layer at a typical growth condition. Thus, 0.4ML of Ga distribution was deduced to be coming from the roughness of the GaInAs surface. Since the Ga composition of the GaInAs layer was about 0.5, when the 0.4ML of Ga distribution was coming from the roughness, the roughness should be 0.8ML (=0.4ML/0.5) and 0.8ML of As distribution should be coming from the roughness. The roughness of the GaInAs surface was estimated to be about 0.8ML by AFM (atomic force microscopy) measurement before the X-ray CTR scattering measurement. The result was consistent with the result of the X-ray CTR scattering analysis.

Concluding the results, formation process of InP/GaInAs interface was able to be described as follows. Just after the growth of the GaInAs layer and before the growth of InP layer, the roughness of the GaInAs surface was about 0.8ML and the surface was covered by adsorbing As atoms of about 4ML-thick. The adsorbing As was able to be desorbed during the growth interruption before the growth of the InP layer. When the growth of the InP layer started, a part of Ga and As atoms in GaInAs layers were incorporated into the InP layer by the exchange of atoms. However the total amount was very small.

4. Conclusions
It has been successfully shown that the X-ray CTR scattering measurement is a good tool to study the formation process of the InP/GaInAs interfaces grown by OMVPE even when the measurement is conducted after the growth. Using the X-ray CTR scattering measurement, the contributions of effects, i.e., surface roughness, adsorption of atoms, and exchange of atoms, on the formation process of InP/GaInAs interface were able to be discussed. The adsorption of As atoms on the GaInAs surface was shown to have the largest contribution on the abruptness of the interface.

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