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Observation of composition in surface monolayers by X-ray scattering spectra caused by crystal truncation and interferences

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X-ray crystal-truncation-rod (CTR) scattering measurements using synchrotron radiation and an imaging plate can reveal the composition of a surface monolayer. Even when the composition is changed in only one atomic layer on the top surface, the X-ray CTR spectrum can change due to the differences in composition. X-ray CTR spectra are greatly enhanced by X-ray interference when a sample is designed properly. In this paper, it is shown by theoretical calculations and experiments for AlAs/GaAs samples grown by MBE that a 1 ML (monolayer)-thick AlAs layer embedded under 10 ML below the surface can enhance the modulation of an X-ray CTR spectrum.

Keywords: X-ray crystal truncation rods; AlAs/GaAs; surfaces; interference.

1. Introduction

When X-ray diffraction from a crystal with a flat surface is measured in detail, a line-like distribution of the X-ray intensity around a Bragg point is observed, which is called crystal-truncation-rod (CTR) scattering. An abrupt truncation of the periodicity of a crystal causes the line-like (rod-like) distribution of the X-ray intensity. Since the structure of overlayers grown on a surface modulates the line-like distributed X-ray intensity, a lot of information about the non-periodic structure of a crystal can be deduced from X-ray CTR scattering (Robinson, 1986; Robinson & Tweet, 1992; Shimura & Harada, 1993).

The X-ray CTR scattering technique is used to investigate the structure and roughness of a surface (Kisker *et al.*, 1992; Lamelas *et al.*, 1994) and interfaces (Robinson *et al.*, 1988; Specht *et al.*, 1991; Hashizume *et al.*, 1992). We have successfully applied the X-ray CTR scattering method to study epitaxically grown semiconductor interfaces and have demonstrated that it is a very powerful technique for revealing atomic-scale interface structures (Takeda *et al.*, 1995; Tabuchi *et al.*, 1995).

In this work, we show that the composition of a surface monolayer can also be revealed by the X-ray CTR scattering measurement. Even when the composition changes in only one atomic layer on top of the surface, the X-ray CTR spectrum is changed and that change reflects the differences in the composition. In addition, it is demonstrated that the change of the X-ray CTR spectra caused by the surface structure can be greatly enhanced by X-ray interference when the layer structures of a crystal are designed properly.

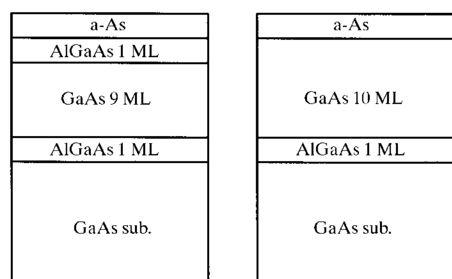

Figure 1
Schematic structures of the samples measured. The surfaces were covered by a-As in order to avoid oxidation of the grown layer.

2. Sample preparation

Two samples, *i.e.* AlAs (1 ML)/GaAs (9 ML)/AlAs (1 ML) on GaAs and GaAs (10 ML)/AlAs (1 ML) on GaAs, were prepared using molecular beam epitaxy (MBE) at 863 K and the surface was covered by a-As (amorphous As) deposited in the same chamber. The MBE growth technique of the AlGaAs/GaAs heteroepitaxial system is one of the most advanced techniques of crystal growth. The structures of interfaces and the compositions of layers in the samples are expected to be formed almost as designed. Therefore, these samples are expected to be very useful for confirming the scope of the X-ray CTR technique. In Fig. 1, the structures of these samples are shown schematically. These samples were capped by the a-As layers in order to avoid oxidation of the grown layer. However, we assumed that these layers did not affect the CTR spectra because the layers were amorphous. Therefore, in this paper, the surface means the top layer of GaAs or AlAs. In a-As/GaAs (10 ML)/AlAs (1 ML), since GaAs (10 ML) is equal to GaAs (1 ML)/GaAs (9 ML), the

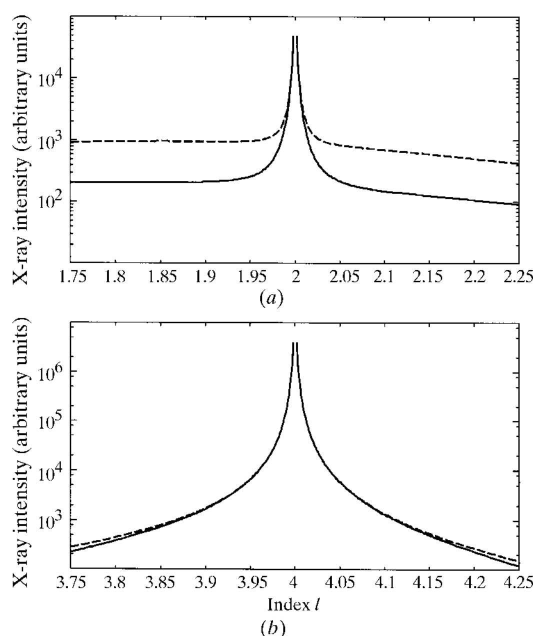

Figure 2
Calculated CTR spectra for (*a*) the 002 and (*b*) the 004 Bragg points. Solid lines and broken lines indicate calculated spectra for the structures with and without 1 ML-thick AlAs on the surface, respectively.

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structure is considered to be a-As/GaAs (1 ML)/GaAs (9 ML)/-AlAs (1 ML). Therefore, the difference between these two samples is only in the composition in the one monolayer (GaAs or AlAs) just under the amorphous As layer. In these samples, 1 ML-thick AlAs layers were embedded under 10 ML below the a-As layer to enhance the effects of X-ray interference.

3. X-ray scattering measurements

The X-ray CTR scattering technique was conducted using synchrotron radiation at beamline $BL6A_2$ of the Photon Factory in the National Laboratory for High Energy Physics at Tsukuba. The wavelength was set at 1.600 Å using a bent Si (111) monochromator. A Weissenberg camera was used to record the X-ray diffraction intensity distribution with an imaging plate as a detector.

4. Theoretical calculations

Fig. 2 shows the theoretically calculated X-ray CTR spectra around (*a*) the 002 and (*b*) the 004 Bragg points of GaAs. The solid lines and broken lines indicate calculated spectra for the structures with and without 1 ML-thick AlAs on the surface, respectively. In the calculation of the spectra shown in Fig. 2, the AlAs layer was assumed not to be embedded in GaAs. As shown in Fig. 2, at least for the AlGaAs/GaAs system, the difference of the surface composition (AlAs or GaAs) can be clearly observed when the X-ray CTR spectra are measured around the 002 Bragg point. However, it is difficult to differentiate between the spectra measured around the 004 Bragg point.

Fig. 3 shows the theoretically calculated X-ray CTR spectrum around the 002 Bragg point of GaAs with the embedded 1 ML-thick AlAs. Solid lines and broken lines indicate the calculated spectra for the structures with and without the 1 ML AlAs on the surface, respectively. The difference, due to whether the 1 ML AlAs is embedded or not embedded, is seen by comparing Fig. 3 with Fig. 2(*a*). Fig. 3 suggests that the embedded 1 ML-thick AlAs layer makes it easier to measure surface composition because the intensity at the peaks is greatly enhanced. When the 1 ML-thick AlAs is not embedded, the shapes of both spectra with and without the 1 ML-thick AlAs on the surface are very similar. Therefore, the absolute intensity of the scattered X-rays around the 002 Bragg point is the only measure of the difference. However, when the 1 ML-thick AlAs is embedded, the shapes of both of the spectra are greatly changed. The modulation of the CTR spectrum of the GaAs capped by AlAs is stronger than that of GaAs without an AlAs cap layer. Therefore, the difference of surface composition can be measured both from the change of the shape of the spectrum and *via* the intensity change itself *i.e.* when the 1 ML-thick AlAs is embedded.

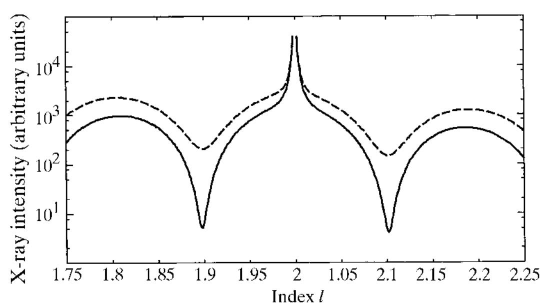

Figure 3
Calculated CTR spectra. Solid lines and broken lines indicate calculated spectra for the structures with and without 1 ML-thick AlAs on the surface, respectively.

5. Experimental results

Fig. 4 shows the measured X-ray CTR spectra of GaAs with and without the 1 ML-thick AlAs on top of the surface. In both samples, the 1 ML-thick AlAs layer was embedded under 10 ML from the surface. As shown in Fig. 4, the measured spectra are very similar to the theoretically calculated ones. This result suggests that X-ray CTR measurements can be a very powerful tool for measuring surface compositions, when the samples are designed carefully.

The experimental results clearly show that the difference of the surface monolayer (AlAs or GaAs) was seen in the X-ray CTR spectra. The intensity of the spectra were enhanced by one order of magnitude *via* interference, without which most of the signals were below noise level. These results also indicate the importance of the embedded 1 ML-thick AlAs layer, and also indicate that the X-ray CTR measurement has the ability to measure the composition change in the surface 1 ML. At the same time, it is important to design the sample structure properly in order to enhance the difference of the spectra caused by the difference of the surface composition.

6. Conclusions

By measuring the AlAs/GaAs samples grown by MBE, it has been demonstrated that the X-ray CTR scattering method using synchrotron radiation and an imaging plate can reveal the

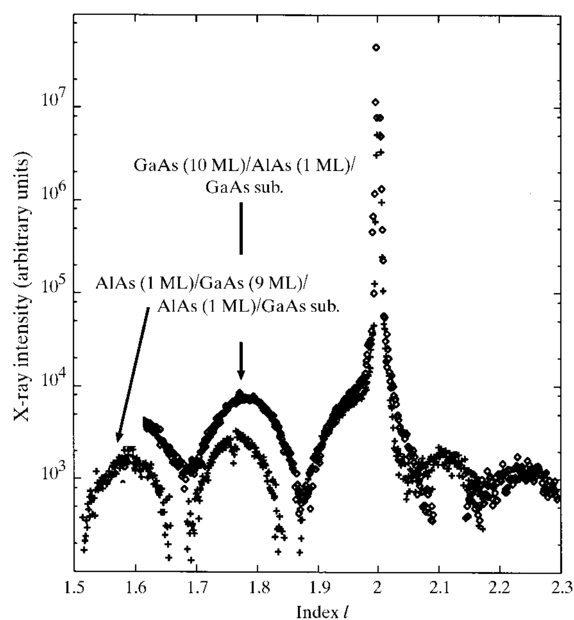

Figure 4
Measured CTR spectra for GaAs (10 ML)/AlAs (1 ML)/GaAs, and AlAs (1 ML)/GaAs (9 ML)/AlAs (1 ML)/GaAs.

composition of a surface monolayer. Even when the composition changes in only one atomic layer on top of the surface, the X-ray CTR spectrum is changed and the change reflects the difference of the composition. The change of the X-ray CTR spectra can be greatly modulated and enhanced by X-ray interference when a sample is designed properly. It was shown by both theoretical calculations and experimental results that a 1 ML-thick AlAs layer embedded at the 11th ML under the surface can enhance the modulation of an X-ray CTR spectrum.

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