Abstract. A ternary Fresnel zone plate (FZP) has been fabricated and installed at the beamline 11XU of SPring-8, in the aim of in situ studies on the growth of semiconductor nanostructures. The FZP is designed for an X-ray energy of 9.5 keV for the first-order diffraction, which is isolated by an order sorting aperture (OSA) inserted 450 mm after the FZP. The focal length of this FZP is 650 mm. The full width at half maximum of the focused beam profile was estimated to be 1.17 μm (H) × 1.38 μm (V) by the dark-field knife-edge scan. Using this FZP, micro-beam diffraction of an as-grown single InAs NW was carried out to show the feasibility of in situ study during growth.

1. Introduction

X-ray diffraction (XRD) has been playing an important role in the characterization of semiconductor nanostructure and provides averaged information, concerning crystal structure, strain, facets, stacking faults, defects and disorientation. The recent achievement of X-ray optics at synchrotron radiation source gives an opportunity to investigate individual nano-sized objects by using single nanostructure diffraction and coherent X-ray diffraction [1-2], which provide unique information of single nanowire (NW) instead of ensemble average of mixed structures of NW with nonuniformity or defects [3]. So far, most measurements of single NWs have been carried out on various grown samples rather than in situ investigation. There is no report to our knowledge of in situ X-ray characterization of single NW.

Beamline 11XU at SPring-8 has a combined setup of a surface x-ray diffractometer and a molecular beam epitaxy (MBE) chamber, which enables in situ studies on the growth of III-V group semiconductors [4]. Recently, we performed in situ X-ray diffraction for real-time monitoring of the crystal structure formation process during self-catalyst GaAs NW growth on Si(111) substrates [5]. The amount of wurtzite (WZ) structure was evaluated as a function of the effective V/III beam flux ratio and its gradient, which may give rise to a crystal structure transition between the WZ structure and the zinc-blend (ZB) structure. The total amount of WZ structure may decrease under low V/III ratios. For in situ investigation of the structure of a single NW, however, a focused X-ray beam is required.

In the present paper, a multilevel Fresnel zone plate (FZP) [6-8] has been developed to focus the incident X-ray beam to 1μm in the high vacuum growth chamber for in situ studies on single nanostructure of III-V group semiconductor during the MBE growth. This ternary type FZP was designed to have a focal length of 650 mm due to the size of the growth chamber, and it has been
installed in the experimental hutch 3 of BL11XU at SPring-8, Hyogo, Japan. Using this FZP, we have carried out a preliminary measurement to evaluate the structure of a single InAs NW on Si(111) substrate. It demonstrated the feasibility of \textit{in situ} study during growth.

2. Experimental

Generally, a nanowire is a nanostructure with a high aspect ratio and diameters on the nanometer scale range from few nm to a few hundreds nm. To carry out \textit{in situ} measurements of individual nanowires, a focused X-ray beam with high intensity is essential and the beam size need to be slightly bigger than the diameter of the object. Moreover, the focal point of the incident beam must be located inside the MBE growth chamber.

A schematic drawing of the experimental setup is shown in Fig. 1. The undulator radiation is monochromatized by a circulated-liquid-nitrogen cooled Si (111) double crystal monochromator. A ternary Fresnel zone plate was installed so that X-rays are focused at the sample position inside the MBE growth chamber located at a distance of 68 m from the light source. A tantalum made slit with an opening of 40 μm in diameter was placed after the monochromator for creating a stable virtual light source, 23.5 m upstream of the zone plate.

![Figure 1](image)

\textbf{Figure 1.} A schematic drawing of the experimental setup for X-ray micro-focusing at BL11XU of SPring-8. CBS: centre beam stop.

To achieve higher intensity, a three-step phase-modulated structure as shown in Fig. 1 was employed. The radii of zone boundaries are designed by

\begin{equation}
 r_{n,l} = \frac{2}{3} f \lambda (3n + l)
\end{equation}

Where λ is the wavelength, \( f \) is the focal length, and n is the zone index number. Each zone is divided into three steps whose index is 0 ≤ \( l \) ≤ 2. Here, \( f \) was designed to be 650 mm for the first-order diffraction at an X-ray Energy of 9.5 keV (\( \lambda = 0.13 \) nm). The diameter of the FZP is 100 μm, and the total zone number and outmost zone width are 19 and 0.5 μm, respectively. The FZP is made of tantalum, fabricated on SiC/SiN membrane deposited on Si wafer. The thickness of Ta steps in each zone are 0 μm, 1.5 μm and 3.1 μm, resulting in the phase shift of 0, \( \pi/2 \) and \( \pi \), respectively. The efficiency of this ternary type FZP is expected to be 48 % as opposed to that of the binary FZP, which is 16 %. To reduce the contribution of the background radiation into diffracted intensity, an Au made 40 μm diameter beam stop was set in front of the FZP. A 35 μm diameter order sorting aperture (OSA) was inserted 200 mm upstream of the focal point in order to select the first-order diffraction and eliminate higher order diffraction.
The evaluation of focused beam profile was carried out by a dark-field knife-edge scan. A gold wire was employed as the knife-edge and mounted on a Ta piece fixed on a molybdenum sample holder, which can be loaded into MBE growth chamber. The diameter of gold wire is 0.3 mm. Figure 2 (a) and 2(b) show the design and picture of the device for knife-edge scan. To measure the beam profiles, the side and lower parts of the wire was aligned perpendicularly to the beam so as to work as the horizontal and the vertical knife-edges, respectively. The scattered X-rays from the gold wire were collected by a NaI detector. As shown in Fig. 3(a) and 3(b), the measured full width at half maximum (FWHM) of the beam profiles is 1.17 μm in the horizontal direction and 1.38 μm in the vertical direction for a focal length of 650 mm.

Figure 2 (a) Schematic drawing of the gold wire mounted on a Ta piece. (b) Picture of the setup. The spot in Fig. 2(a) represents the focal point.

Figure 3 Measured beam profiles of focused beam using dark-field knife-edge scan. (a) Horizontal beam profile and (b) vertical beam profile.

3. Micro-beam diffraction on single InAs nanowire
The X-ray micro-beam focusing system was applied to evaluate the structure of a single InAs NW at 9.5 keV. The sample used was an MBE-grown InAs nanowires on a patterned Si(111) substrate covered by 30 nm SiO₂ layer. The distance between neighbouring wires was estimated to be about 5 μm and the average diameter of InAs NWs was 150 nm as determined by scanning electron microscopy (SEM). In order to localize a position of single InAs NW, an 20×20 μm² region in the sample centre area was scanned in a mesh using InAs 111 reflection as a probe signal, with a step of 5 μm in the horizontal direction and 1μm in vertical direction. The diffracted X-rays were measured with a two dimensional Pilatus 100K detector placed at a distance of 700 mm from the sample.
A single nanowire was selected by moving the sample stages to one of the positions indicated by the mesh scan. At this position, a reciprocal space mapping (RSM) was carried out by rotating the sample around the InAs 111 Bragg point $\theta_B$ with a step of 0.014°. Figure 4 shows a cross section at the (111) plane of the obtained 3D RSM. The obtained high-quality diffraction pattern with fringes demonstrated the measured object was a single NW whose diameter was estimated to be 157 nm from the spacing between interference fringes. The six-fold symmetry of the diffraction pattern indicates the hexagonal shape of the measured nanowire. For in situ measurement, pre-deposited catalyst nanoparticles or holes opening on the patterned substrate can be used to localize possible positions of individual NWs before the MBE growth. In the case of self-assembled NWs, an initial growth of small amount of nanowires will help to select a position of single nanowire prior to in situ measurement.

4. Conclusion
We developed a ternary FZP for in situ studies of individual III-V compound nanowires during MBE growth. The beam size at the focal point was 1.17 μm in the horizontal direction and 1.38 μm in the vertical direction as measured by dark-field knife-edge scan. Diffraction from a single InAs NW was successfully measured.

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