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X-ray diffraction study of evolution of polytypes in Au-assisted GaAs nanowires

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Abstract

The polytypism of GaAs nanowires was investigated by in situ X-ray diffraction using a molecular-beam epitaxy chamber combined with an X-ray diffractometer. The growth of nanowries was found to start with the formation of zincblende structure, followed by the growth of the wurtzite structure. The wurtzite structure tended to form at a low growth temperature and a high growth rate.

1. Introduction

Nanowires of III-V semiconductors are known to show poytypes including the 6H, 4H and wurtzite (2H) structure in addition to the zincblende (3C) structure that is the most stable in the bulk. Since the electronic and optical properties of nanowrires are sensitive to the crystal structure, the control of polytypes is required for device applications of nanowires. For this purpose, understanding of the growth mechanism of nanowires is important. In this paper, we present in situ X-ray diffraction results revealing the transition of the dominantly growing crystal structure under different growth conditions.

2. Experimental

Sample preparation

Experiments were performed at the synchrotron radiation beamline 11XU at SPring-8 using molecular beam epitaxy chamber integrated with a surface x-ray diffractometer [1]. We employed the vapor-liquid-solid method [2, 3] to grow GaAs nanowires on GaAs(111)B. After desorbing native oxide layers, 1.8-nm-thick Au was deposited at substrate temperature of 560℃. Then the substrate was cooled to temperatures ranging from 430 to 480°C for GaAs nanowire growth. By a separate X-ray diffraction measurement, it was confirmed that Au-Ga alloy remained melted in this temperature range. The deposition rate of Ga corresponded to the two-dimensional growth rate of 200 nm/h in the homoepitaxial growth of Ga(001).

X-Ray diffraction measurements

X-ray diffraction measurements were performed under in situ conditions while the nanowire growth was temporarily being interrupted. An X-ray wavelength of 0.124 nm was selected using a liquid-nitrogen-cooled Si(111) double crystal monochromator. We measured the intensity distribution along the reciprocal rod, (1,-1, L), where the diffraction indices are represented by the hexagonal coordinates based on the unit cell defined by $a=[1/2,0,-1/2]_{cubic}$, $b=[0,-1/2,-1/2]_{cubic}$, and $c=[-1,-1,-1]_{cubic}$. Diffracted X-rays were collected by a charge coupled device detector, which was placed at a distance of 700 mm from the sample. By virtue of the two-dimensional imaging of this detector, the diffraction signal from the crystal and the background including the thermal diffuse scattering can be separated. At each point of L, the diffracted intensity was integrated over the diffraction peak and the background intensity was subtracted.

3. Results

Growth temperature dependence

Figures 1 to 3 show the evolution of the X-ray diffraction profiles along the (1,-1) rod during the GaAs nanowire growth at substrate temperatures of 430°C, 455°C and 480°C, respectively. The peaks at L=2 and 5 correspond to the 3C structure of the GaAs substrate. At 480°C, the GaAs nanowire growth was initiated by the formation of the 3C nanowires. The growth of the 3C structure continued until the deposition amount reached 6.7 nm. After this thickness 2H peaks were found to show up at L=1.5, 3.0 and 4.5. A closer look at the diffraction profile shows additional faint peaks attributable to the 4H structure at L=2.25 and 3.75. At a lower growth temperature, it was found that the growth mode transition from 3C to 2H took place at a smaller GaAs thickness. The 2H peaks appeared earlier at a temperature of 455°C. At 430°C, the 2H structure was observed from the beginning of the growth.

III to V ratio dependence

The deposition rate of Ga was found to influence on the crystal structure of nanowires. Figures 4 (a) and (b) show the X-ray diffraction profiles of GaAs nanowires corresponding to the Ga depsotion rate of 6 ML/min and 24 ML/min, respectively. The total growth time was 15 min. For the GaAs nanowires grown at the lower growth rate, quarter-order peaks at L = 1.75, 2.25 and 3.75 are clearly observed. These peaks are indicative of the well-ordered 4H structure. At a higher growth rate of GaAs, the 4H peaks were found to diminish as shown by the curve (b) in Fig. 3. In addition, the half-order peak at L = 1.5, 3.0 and 4.5 becomes broader. This is due to the transition from 4H to 2H.

4. Conclusions

The polytypes in GaAs nanowires were investigated by in situ X-ray diffraction. Zincblende nanowires were found to be formed at the initial stage of the growth, followed by the formation of the wurtzite structure. Wurtzite nanowires were preferentially formed at a low growth temperature and at a high growth rate.

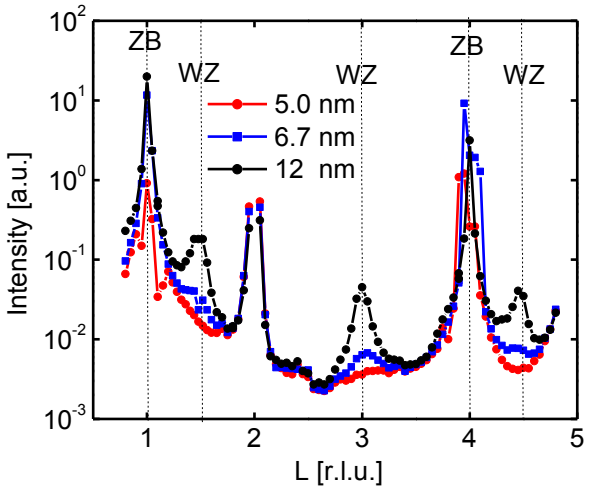

Fig. 1 Evolution of X-ray diffraction profiles along (1, -1, L) during the growth of GaAs nanowires at a substrate temperature of 480°C.

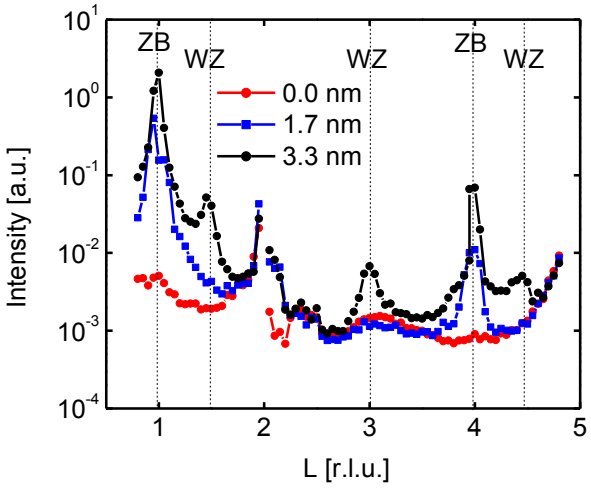

Fig. 2 X-ray diffraction profiles along (1, -1, L) during the GaAs nanowire growth at 455°C.

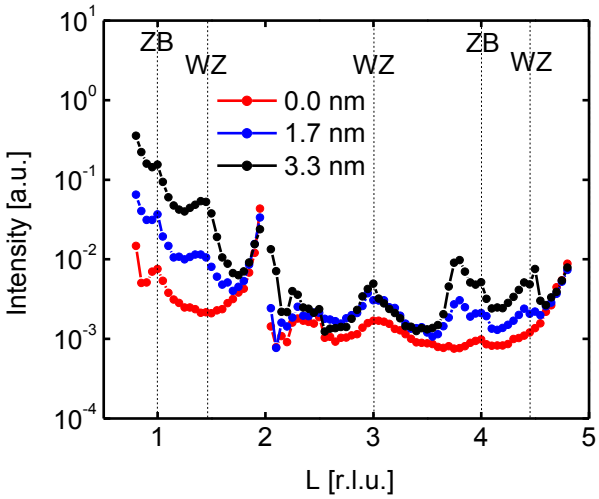

Fig. 3 X-ray diffraction profiles along (1, -1, L) during the GaAs nanowire growth at 430°C.

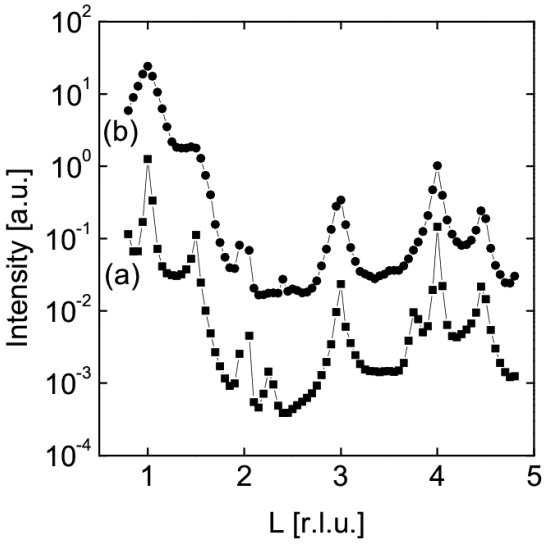

Fig. 4 X-ray diffraction profiles along (1, -1, L) from the GaAs nanowires grown at a deposition rate of 6 ML/min (a) and 24 ML/min (b).

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