

ELEMENT-SPECIFIC SURFACE X-RAY DIFFRACTION STUDY OF GaAs SURFACES

Gallium arsenide is a semiconductor showing excellent performance in optoelectronic and high-frequency devices. It plays a critical role in today's highly networked society, being used in cell phones, satellite communication and laser diodes. For these applications, atomically controlled device structures are grown by molecular-beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD). A detailed understanding of surface structures under growth conditions is an important basis for these techniques.

Atomic arrangements on a semiconductor surface often differ from those of the ideal bulk structure. Firstly, atoms are displaced from the bulk positions to reduce the number of dangling bonds at the surface. The resultant surface reconstruction involves several atomic layers that are below the surface. Secondly, through the exchange of atoms between the substrate and the environment, chemical compositions are allowed to deviate from that in bulk in the case of compound semiconductors. This flexibility in stoichiometry results in a number of variations of surface reconstructions. For the full determination of a surface reconstruction, therefore, both the positions and the species of surface atoms need to be determined. Although electron diffraction analysis, which is most widely used for surface structure analysis, is capable of completing this task in principle, it is very difficult to differentiate atoms with similar atomic numbers such as Ga and As. Scanning tunneling microscopy cannot differentiate atomic species either. Element-specific techniques, such as ion scattering, photoemission spectroscopy and the analysis of X-ray fluorescence excited by electrons, have trouble distinguishing the signals from the surface and the substrate.

Figure 1 shows the GaAs(001)-c(4×4) structure investigated in the present work. This surface was used as a template for the growth of InAs quantum dots or dilute magnetic semiconductors and thus has been attracting much interest recently. Because this surface was observed under As-rich conditions, it was initially considered that all the atoms in the topmost layer were As atoms [2]. Later on, however, a new structure model in which half of the top-layer atoms are considered to be Ga atoms was proposed [3]. Experimentally, the claim of the presence of Ga was based on the presence of an asymmetric structure with respect to the [1 -1 0] axis accompanied by the

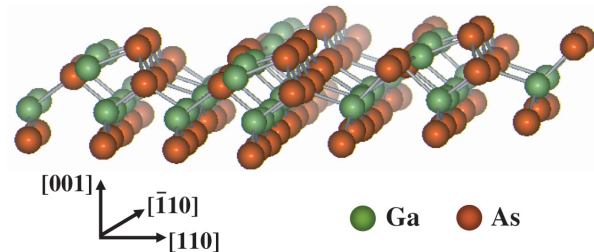


Fig. 1. Ball-and-stick model of GaAs(001)-c(4×4).

buckling of surface dimers. However, asymmetric dimers are widely observed structures even for single-element semiconductors, such as Si(001) and Ge(001). Direct evidence of Ga-As heterodimer formation was not available at the time. Thus, we applied element-specific surface X-ray diffraction analysis of GaAs(001)-c(4×4) for surface structure determination that specified atomic species [1].

For surface studies under crystal growth conditions, we have developed an X-ray diffractometer integrated with an MBE chamber at beamline **BL11XU** [4]. The intense X-rays of SPring-8 have speeded up the measurements of surface X-ray diffraction, which is weaker than the diffraction from bulk crystals by several orders of magnitude. Moreover, the wide spectrum of SPring-8 X-rays has enabled the determination of the species of surface atoms as well as their coordinates.

Our analysis was carried out in two steps. The first step was determining atomic positions by conventional surface X-ray diffraction measurements at an X-ray energy of 10 keV. By combining in-plane and out-of-plane measurements, we determined the three-dimensional coordinates of the atoms up to the sixth layer from the surface. As shown in Fig. 2, the simulation based on the asymmetric-dimer model fits the measurements better than the symmetric-dimer model. The experimentally determined coordinates showed very good agreement with those obtained using a first-principles calculation [5]. Still, it was unclear whether the dimers are As-As homodimers or Ga-As heterodimers. Even when both atoms in the surface dimer were assumed to be As, a similarly result was obtained because the atomic numbers of Ga and As are very close, 31 and 33, respectively. Thus, up until this stage, we had just confirmed the formation of asymmetric dimers rather than that of heterodimers. To resolve this uncertainty, we

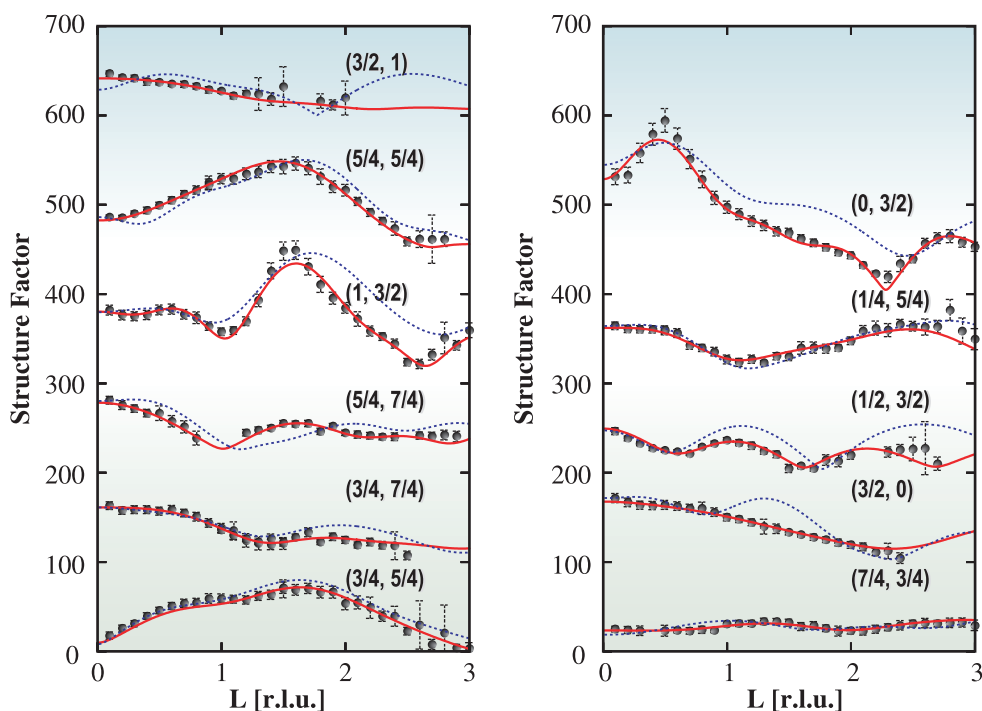


Fig. 2. Comparison of measured (filled black circles) and simulated (red and blue lines) fractional-order rods. The simulation based on the asymmetric dimer model (solid red lines) give better fitting than the symmetric dimer model (dashed blue lines).

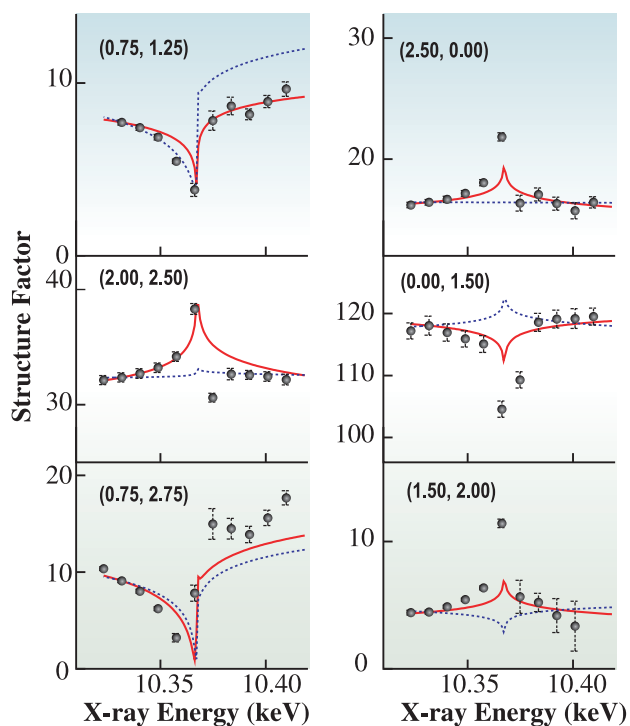


Fig. 3. X-ray energy dispersion of the structure factors. The circles are observed structure factors. The red and blue lines represent the results from calculations when Ga-As and As-As dimers are assumed, respectively.

measured the variation in X-ray diffraction intensity as a function of X-ray energy. As shown in Fig. 3, the experimental results (filled circles) agree better with the model in which the topmost layer consists of both gallium and arsenic (red lines) than the counterpart in which only arsenic is present on the surface (blue lines).

In summary, we have investigated the structure of GaAs(001)-c(4×4) by element-specific surface X-ray diffraction analysis. The formation of Ga-As surface dimers in this surface has been confirmed using the anomalous scattering of X-rays. This study has provided direct experimental evidence of the presence of Ga-As dimers in GaAs(001)-c(4×4).

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