

# **Characterization of Columnar InAs/GaAs Quantum-Dot Structures Using Grazing Incidence X-ray Diffraction**

Kohki Mukai\* and Yuuta Kimura

Department of Mechanical Engineering and Materials Science,

Yokohama National University,

79-5 Tokiwadai, Hodogaya-ku, Yokohama 240-8501, Japan

\*E-mail address: mukai@ynu.ac.jp

Grazing incidence X-ray diffraction (GIXD) measurement using equipment available for laboratories was realized for the evaluation of columnar InAs/GaAs quantum dots (QDs). The QDs were grown by direct stacking of small self-assembled islands in the growth direction, and we prepared some samples by changing the number of units stacked. Using an imaging plate for a detector, we successfully observed that QD-related signals appeared around a GaAs(220) peak. The QD signals extended in the direction of the  $2\theta$  axis, and the extension increased with increasing number of stacks in the columnar QD growths. The results of the measurements were evaluated by a simulation based on the finite element method. Using a simple model that divides the

QDs into an InAs columnar core and an InGaAs transition domain, we found that the core was expanded according to the increase in the stacking number. We also found that the lattice constant increased almost uniformly toward the upper part inside the QD. GIXD evaluation in laboratories will enhance the speed of the development of QD devices.

## 1. Introduction

Quantum dots (QDs) are very attractive optical device components. Semiconductor QD lasers have many advantages over conventional quantum-well lasers, such as the stability of lasing threshold and output power with temperature changes, low power consumption, and high-speed direct-modulation operation.<sup>1)</sup> The self-assembled QDs have thus far been eagerly studied all over the world and have been markedly improved in homogeneity, emission wavelength, multilayering, and controllability of shape. After room temperature operation of a practical QD laser was achieved,<sup>2)</sup> the predicted performances have been proved.<sup>3)</sup> Commercialization of QD lasers will be realized soon. In addition, a QD semiconductor optical amplifier (SOA) is another noteworthy recent research subject.<sup>4,5)</sup> A single photon emitter with QD is also achieving remarkable success for the future applications to quantum communication.<sup>6-8)</sup>

For the development of QD optical devices, it is very important to correctly control the QD's internal structure which directly determines the performance of carriers. In particular, the structural symmetry of QDs affects the polarization characteristics in a QD-SOA<sup>9,10)</sup> and the generation of an entangled photon pair as a photon source.<sup>11-13)</sup> A method to control the carrier

confinement direction within the QD structure has been developed by stacking small QDs directly in the growth direction. These so-called columnar QDs have succeeded in improving the homogeneity in energy<sup>14)</sup> and the symmetry of the QD structure.<sup>15)</sup>

For further development of structure controllability, it is important to acquire internal information about QDs in detail. Cross-sectional scanning tunneling microscopy is a powerful tool for acquiring information on QD structure with atomic accuracy,<sup>16)</sup> but it is a destructive method of inspection and requires expensive special sample preparation. Grazing incidence X-ray diffraction (GIXD) is an effective nondestructive evaluation method that can clarify the strain and composition distribution inside QDs at nm resolution. However, a synchrotron radiation X-ray source has been required for the measurement,<sup>17-19)</sup> and therefore, the speed of response was not sufficient for use in actual device development.

In this work, we succeeded in performing GIXD measurements using equipment available for laboratories and characterized columnar InAs/GaAs QD structures. Evaluation of the internal structure of a QD was performed by comparing the results of the measurements with those of simulations.

## 2. Experimental Methods

Columnar QD samples were prepared by solid source molecular beam epitaxy (MBE). The samples were grown on (001)-GaAs substrates at 510 °C. To form a single columnar QD, 0.7-monolayers of InAs QDs were directly stacked with 3-monolayer intervals of GaAs layers after 1.8-monolayers of InAs were supplied [Fig. 1(a)]. Since multiple stacking growth is carried out, multiple wetting layers are formed on the circumference of the columnar QDs. We prepared four samples where the number of InAs stacking units varied from 9 to 32.

The GIXD measurements were performed to evaluate the columnar QD structure. Figure 2 shows the experimental setup. A "PANalytical X'Pert PRO MRD" system was used as the X-ray source. An incident X-ray beam was located at the very small angle near the total-internal-reflection angle to a (001) plane surface. Most of the incident beam was reflected at the surface plane. We set the samples so that diffraction might occur in the (220) plane. Therefore, the obtained data represents lattice information in the direction parallel to the surface. The diffracted X-ray beam was irradiated on an imaging plate (IP) and detected as two-dimensional information.

### 3. Results and Discussion

Figure 3 shows the signal images on IPs of four samples for GIXD measurements. The  $n$  denotes the number of small QDs stacked to form columnar QDs. The deepest signal is GaAs(220) diffraction. Since sample sizes were not the same, absolute intensity varied. The signal at the upper right of the GaAs(220) peak is the diffraction from the columnar QDs. The signal extends in the  $2\theta$  direction, and the extension increases as the number of QDs stacked,  $n$ , increases.

The differences among samples were clearly indicated by converting the data from the IPs into rocking curves. Figure 4 shows the relationship between the linear diffraction intensity and the diffraction angle detected by the IPs. The diffraction intensity is the value integrated in the  $\alpha_f$  range containing signals from the QDs. Therefore, the GaAs(220) peak is not necessarily the highest in the rocking curves. Diffraction peaks of QDs were observed at a smaller angle to the GaAs peak and spread to the low-angle side according to the increase of  $n$ . Figure 5 shows the full width at half maximum (FWHM) and center positions of the QD-related peaks as a function of  $n$ . It clearly appears that the FWHM increased and peak position decreased as the stacking number increased, suggesting a systematic change in sample structure.

We simulated the rocking curves to analyze the results of X-ray diffraction. In the simulation, we first calculated the three-dimensional strain-field distribution based on the compositional distribution inside and outside the QDs using the finite element method.<sup>20)</sup> For simplicity, the columnar QD was modeled as shown in Fig. 1(b), where the QD has an InAs core and a surrounding cylindrical InGaAs transition domain. Note that near the edge of the columnar QD the upper and lower small QDs do not touch directly; this is the transition domain between the InAs core and the surrounding multiple wetting layer. The diameter of the QD was assumed to be 15 nm. The multiple wetting layer was neglected. Under the conditions of minimizing strain energy, we determined the lattice constant distribution. Then, rocking curves were calculated for the QD structures with dynamical theory based on the structural parameters.

Indium composition,  $x$ , and width of the transition domain,  $d$ , were considered fitting parameters when comparing the experimental results with the calculated ones. During the adjustment of parameters, values from 1.5 to 5.0 nm were examined as the width of the transition domain, and values from 0 to 1.0 were examined for the indium composition of the transition domain. The relationship between the fitting parameters and the diffraction peak was checked in

advance for reference. Figures 6(a)-6(d) show the results. In the calculations,  $d = 2.5$  was assumed for Figs. 6(a) and 6(b), and  $x = 0.5$  was assumed for Figs. 6(c) and 6(d). A common tendency was found for all QD stacking numbers. As the indium composition of the transition domain increases, FWHM increases and the peak position moves to smaller angles. As the width of the transition domain increases, FWHM decreases and the peak position moves to larger angles. These findings helped us in fitting the results of GIXD measurements. Figure 7 shows the rocking curves obtained from calculations, in which the parameters of the transition domain were chosen so that the experimental results of the QD diffraction were reproduced. In the calculations, only the X-ray diffraction from QD and its neighbors was considered in the kinematical scheme, and therefore the diffraction from the GaAs substrate and the peak broadening due to the dynamical scheme were not considered in the figures. The good agreement between the calculations and the measurements shown in Fig. 4 was obtained for all samples. From the trend given to the calculations by the parameters, we consider that the result obtained here was the best fit. As a consequence, we found that the indium composition of the transition domain increased from 0.5 to 0.9 and the width of the transition domain decreased from 5 to 1.5 nm in connection with the increase of the number of stacking units,  $n$ , from 9 to 32. The results suggest that the InAs core grew as stacking number increased.



The internal structure of QD samples was investigated by analyzing the QD-related diffraction in detail. We applied the method reported by Kegel et al.<sup>17)</sup> to the sample for  $n = 32$ . The height in the growth direction can be determined from the incidence angle,  $\alpha_i$ . We sliced the data on IPs into many rocking curves using the reflection angle,  $\alpha_f$ . Assuming  $\alpha_i = \alpha_f$ , the lattice constant distribution was obtained from the rocking curves as a function of height.

Figure 8 shows the in-plane lattice constant distribution in the QDs. Owing to the insufficient resolution of the IP analyzer, the distribution is a step-like function. This figure suggests that the in-plane lattice constant increases monotonically toward the upper part of the QD. The lattice constant was directly determined from the position of the X-ray diffraction peak, and it indicates that the lattice constant of the upper part of the QD was larger than the lattice constant of free-standing InAs. This means that crystal distortion was distributed in three dimensions so that the form of the crystalline lattice became flat in the upper part. It should be noted that the growth conditions were not changed at all during the growth of columnar QDs. Therefore, the lattice constant distribution in the growth direction is probably caused by nonintentional atomic movements during growth, such as interdiffusion and surface reconstruction.<sup>21)</sup> Another explanation is the relaxation of lattice distortion at the top of

the QD. This could be unusual behavior caused by the introduction of crystal dislocations with a very high aspect ratio inside the QD. The contribution of these effects will be clarified by transmission electron microscopy and the analysis of the indium composition distribution. X-ray analysis in three dimensions will also aid in dividing the information on lattice size into information on composition and strain. These analyses are left for a future study.

#### **4. Conclusions**

We realized GIXD measurements with equipment available for laboratories and performed structural evaluations on columnar QDs. Using an IP for a detector, we successfully observed that QD-related signals appeared around the GaAs(220) peak. The signals extended in the  $2\theta$  direction, and the extension varied with the number of small QD stacking units that formed the columnar QD. We simulated the results of X-ray diffraction by calculating the distribution of strain fields in the samples based on the finite element method. Assuming a simple model that divides the columnar QDs into an InAs core and an InGaAs transition domain, we found that the core expanded according to the increase in the number of QD stacking units. We also found that the lattice constant increased almost uniformly toward the upper part of the inside of the QD. The GIXD measurement with equipment available for laboratories will be a

powerful tool for the development of QD devices.

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### **Figure captions**

Figure 1. (a) Schematic of columnar QD in cross section, and (b) model of QD used in the simulation.

Figure 2. Experimental setup for GIXD measurement.

Figure 3. (Color online) Imaging plate around GaAs(220) signals in four samples.

Figure 4. Measured rocking curves of four samples with various numbers of stacking units.

Figure 5. FWHM and peak position of diffraction peaks as a function of the number of stacking units.

Figure 6. (Color online) Calculated (a) FWHM and (b) peak position of diffraction peaks as a function of the indium composition of the transition domain. Calculated (c) FWHM and (d) peak position as a function of the width of the transition domain.

Figure 7. Calculated rocking curves of four samples. In the calculations, the parameters of the transition domain were chosen so that the experimental results were explained by our assumptions.

Figure 8. Lattice constant distribution for a QD of  $n = 32$  as a function of height.

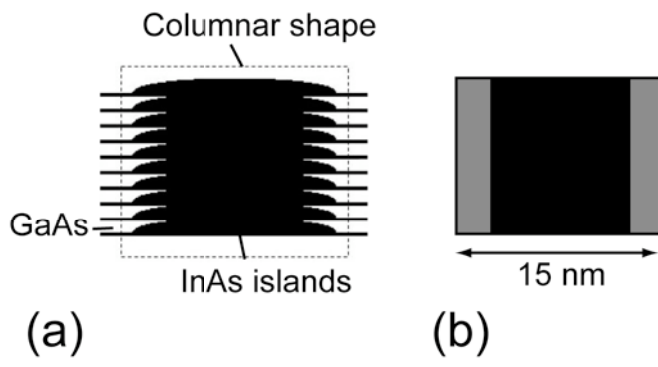


Figure 1



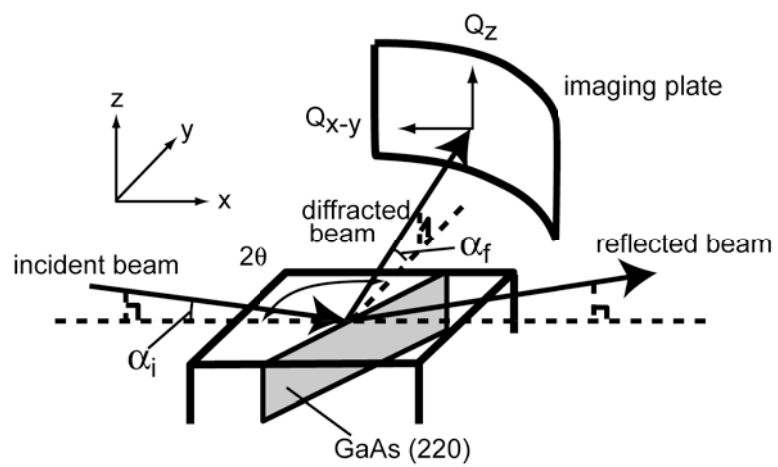


Figure 2

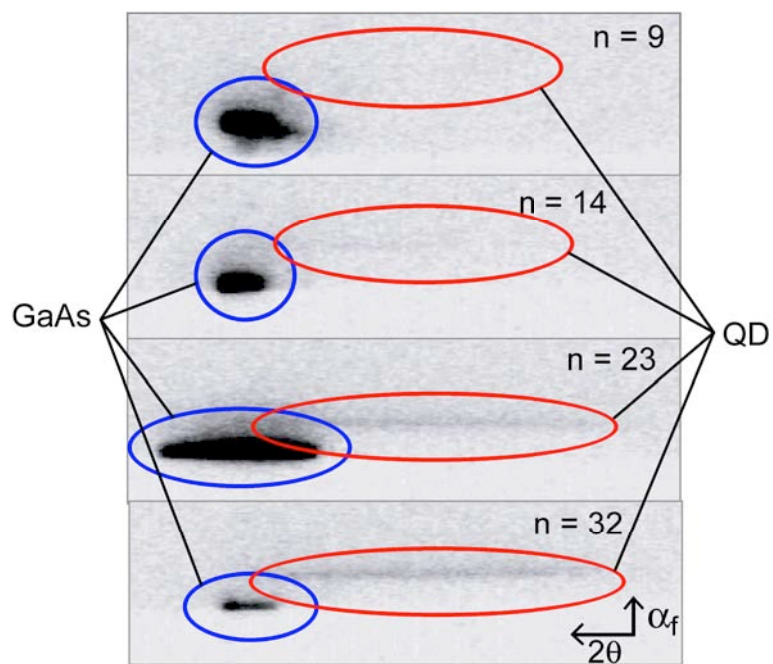


Figure 3

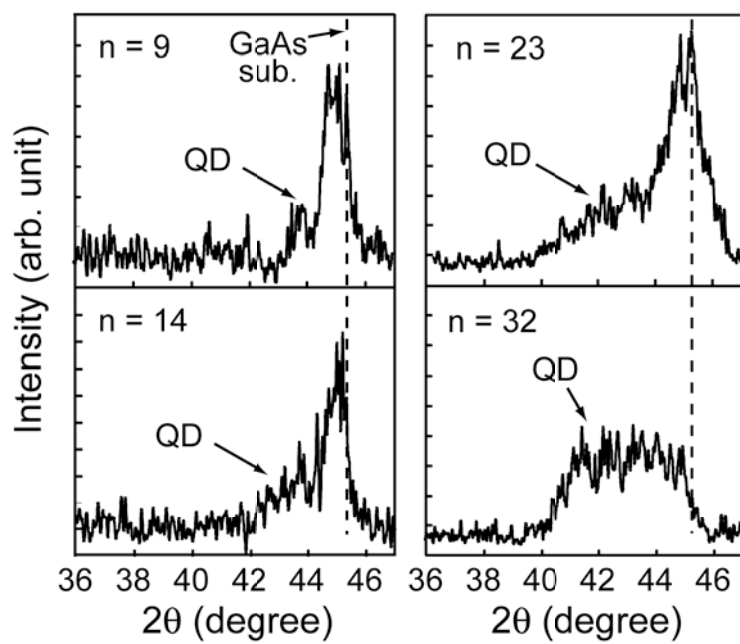


Figure 4

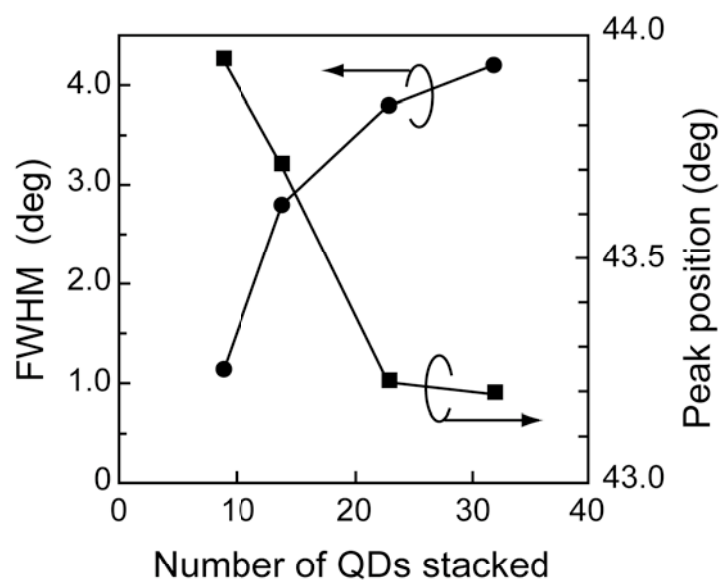


Figure 5

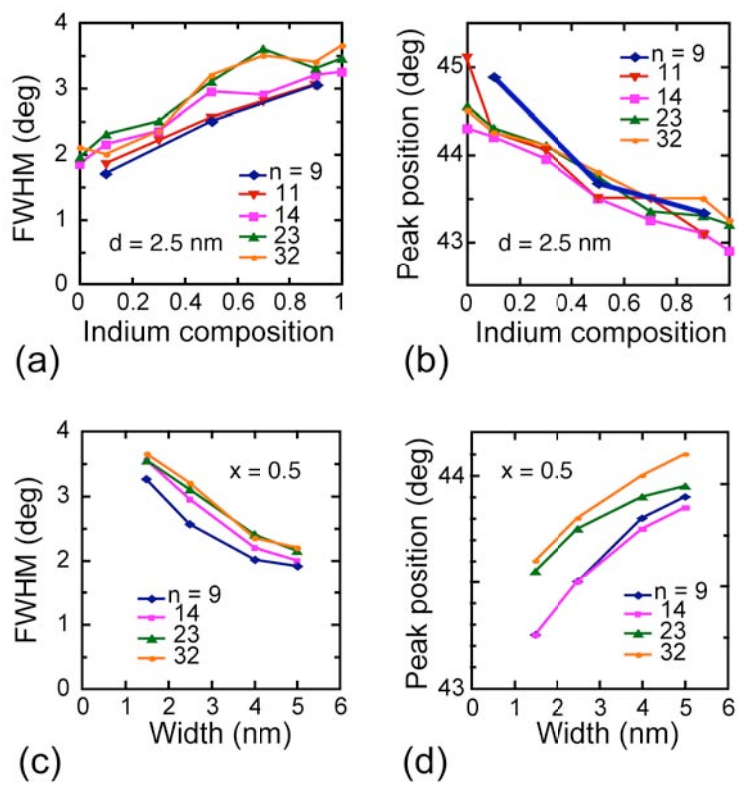


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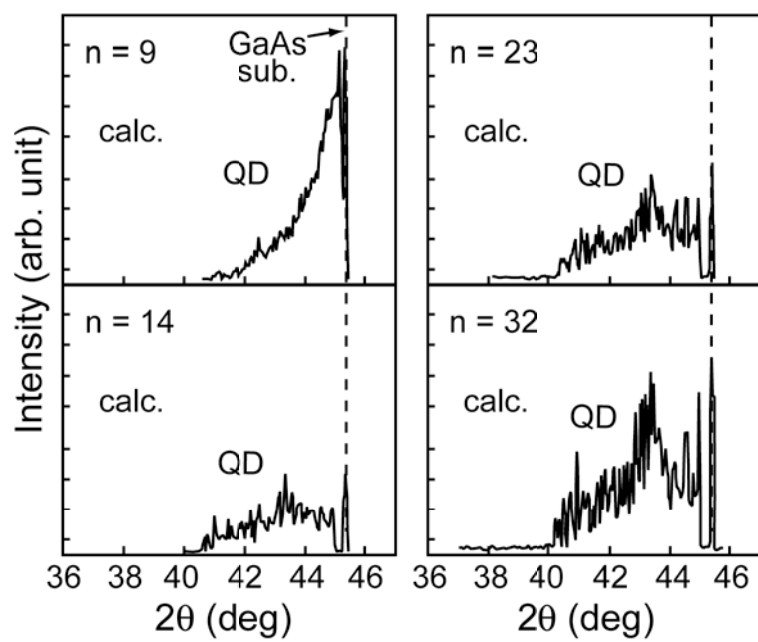


Figure 7

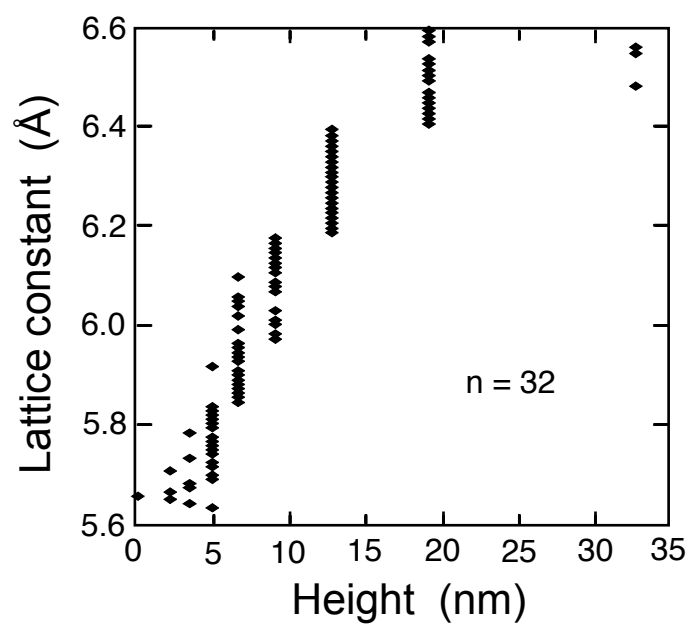


Figure 8