Lattice deformation and interdiffusion of InAs quantum dots on GaAs(100)

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InAs quantum dots (QDs) on GaAs(100) grown by molecular-beam epitaxy were structurally characterized by ion channeling. Lattice deformation of the InAs QDs and diffusion of Ga atoms into InAs QDs were clearly observed to depend strongly on the InAs coverage. It was revealed that the diffusion is significantly enhanced when the InAs coverage is changed from 1.53 to 1.71 monolayer. During this change, lattice deformation was reduced while the average size (base diameter) of dots was decreased. These phenomena suggest that some growth process change occurred. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1328777]

I. INTRODUCTION

Recently, a great interest in the self-formation of three dimensional quantum dots (QDs) has arisen.[1–7] They are grown by Stranski–Krastanow growth mode in heteroepitaxial material systems having a large lattice mismatch. In particular, InAs QDs on GaAs(100) have been challenged to be used for potential optoelectronic devices.[8–10] Many studies on the optical and electrical properties have been done.[5,10–13] In our previous report,[14] an accurate value of InAs coverage was determined and diffusion of Ga atoms into InAs QDs was observed by using Rutherford backscattering (RBS). Joyce *et al.* reported the Ga diffusion, and its dependence on the growth temperature.[15] However, the influence of the diffusion on lattice deformation of the system is not yet fully understood although the influence is important to describe a conclusive picture of the self-assembled growth process. Moreover, the accurate value of the InAs coverage for the sample having a thick GaAs cap has not been determined although the coverage is expected to have a strong influence on the diffusion on lattice deformation. In this article, we carefully determined the InAs coverage for the capped samples, and we investigated the dependence of the crystallographic structure of InAs QDs on the InAs coverage by using ion channeling observed by RBS and particle induced x-ray emission (PIXE). The channeling investigation is highly sensitive to atomic arrangements.[16,17]

II. EXPERIMENT

All samples contained a single InAs layer, and were grown by molecular-beam epitaxy (MBE). A 500 nm thick GaAs buffer layer was first grown at 655 °C with the growth rate of 800 nm/h. Subsequently, InAs was grown at 510 °C. Moreover, a 150 nm thick GaAs layer was overgrown as a cap layer. Nominal thicknesses of the InAs layer were estimated by the growth rate of the order of 0.1 monolayer (ML)/s. Samples without GaAs cap layers were also prepared to investigate the diffusion.

A standard experimental arrangement for ion channeling was used with Van de Graaff-type ion accelerator at Hokkaido University, which has a three-axis goniometer. A Si annular solid state detector (scattering angle: 174°) was used for RBS, and a Si(Li) solid state detector was also used for PIXE. 1.00 MeV H^+ ions were mainly used as a probe beam to investigate ⟨100⟩ channeling properties. The beam spot and the divergence were 1.0 mm and $<0.03°$, respectively.

To evaluate lattice deformation, we used the normalized minimum yield, $\chi_{\min}$, which defined as a ratio of aligned yields to random ones. From this ratio, we determined the degree of lattice deformation. Here, the lattice deformation is defined as the lateral displacement of atoms from the GaAs host lattice.

Before characterizing the crystallographic structure, we determined the accurate value of the InAs coverage of samples having a GaAs cap layer. The process is the following (Fig. 1). In the RBS spectrum, the InAs of an uncapped sample formed a separate peak (see, for example, Fig. 7). From the integral of this peak, we could determine the absolute value of the average InAs coverage. Here, we used the random yield of GaAs to calibrate the RBS signal of In.[14] At the same time, we detected the PIXE signal of In from the uncapped sample, and calibrated this PIXE signal by the absolute value of InAs coverage obtained. For the capped sample, however, we cannot separate the InAs signal from the RBS spectrum. Instead, we obtained the PIXE signal which decayed about 6% by the absorption of the 150 nm GaAs cap layer. By correcting the decay, we estimated the PIXE signal without the cap layer which was already calibrated to give the absolute InAs coverage. The results are shown in Table I. The accuracy is estimated to be ±0.02 ML.

III. RESULTS AND DISCUSSION

Figure 2 shows typical RBS/channeling spectra for the 150 nm thick GaAs capped sample. Open circle indicates

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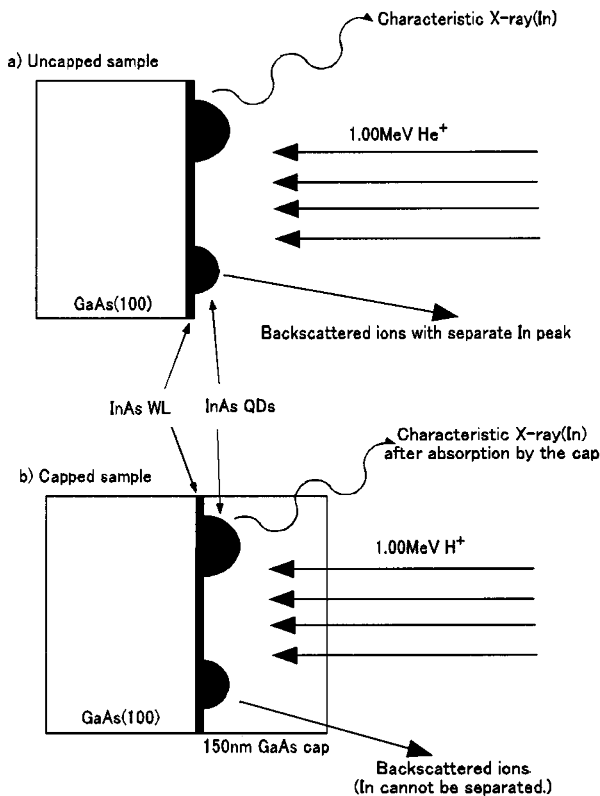

FIG. 1. Schematic drawing of measuring the accurate value of InAs coverage.

⟨100⟩ aligned spectrum. Figure 3 shows the summarized results of RBS/channeling. In brief, $\chi_{\min}$ of RBS tends to increase as the InAs coverage increases. This generally suggests that a larger InAs coverage makes the lattice more deformed. Strictly speaking, however, the value of $\chi_{\min}$ for sample C (InAs coverage; 1.78 ML) was slightly smaller than that of sample B(1.56 ML). In order to obtain separate information on In, Ga, and As, PIXE/channeling experiments were carried out with 1.00 MeV H^+ ions. Figure 4 shows typical PIXE spectra for the sample having a 150 nm thick GaAs cap layer. Figure 5 shows $\chi_{\min}$'s of PIXE as a function of the InAs coverage. The RBS of Fig. 3 is also shown in Fig. 5. Behavior of $\chi_{\min}$ for As is almost the same as that for RBS, and those values of $\chi_{\min}$ for sample C were slightly smaller than that of sample B. The magnitude of $\chi_{\min}$ for In changed drastically with varying InAs coverage. The value of $\chi_{\min}$ for In of sample A (1.02 ML) was very close to that for RBS. This indicates that the InAs layer is pseudomorphically grown on the GaAs substrate, which means that InAs is grown two dimensionally (2D). In fact, no dots were suggested to be formed by reflected high energy electron diffraction during the growth of this sample. The value of $\chi_{\min}$ for

TABLE I. Nominal and accurate InAs coverage (ML) (A–D: capped, E–J: uncapped).

	A	B	C	D	E	F	G	H	I	J
Nominal	1.1	1.6	1.8	2.3	1.3	1.6	1.8	1.8	2.1	2.6
Accurate	1.02	1.56	1.78	2.35	1.32	1.53	1.71	1.73	2.23	2.58

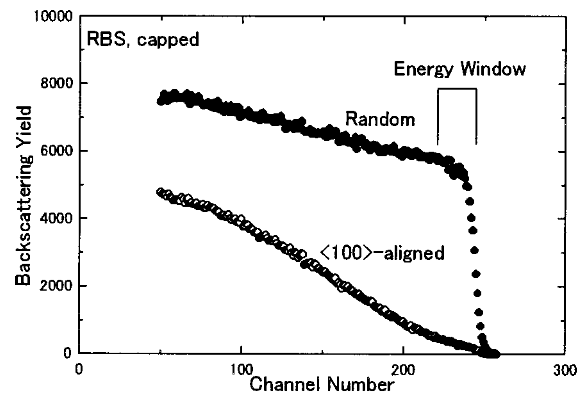

FIG. 2. Typical RBS channeling spectra for a 150 nm thick GaAs capped sample.

In drastically increased, and showed the maximum at sample B. This indicates that the InAs growth mode shifted from 2D to three-dimensional (3D) and lattice deformation of InAs QDs was observed. The critical coverage for 2D–3D growth transition, θ_c, has been reported to be around 1.5 ML.[4–7,9] For sample C, the magnitude of $\chi_{\min}$ for In suddenly decreased. It should be noted that this remarkable difference appeared with a growth-time difference of only two seconds. Beyond this, $\chi_{\min}$ gradually increased when the coverage was changed from 1.78 to 2.35 ML (sample D). Since $\chi_{\min}$ for In is expected to reflect the lattice deformation of InAs QDs, lattice deformation of the InAs QDs strongly depends on the InAs coverage. While $\chi_{\min}$'s for RBS, As, and In in sample C are smaller than those in sample B, slight increase of $\chi_{\min}$ for Ga is observed at this point.

Figure 6 shows the variation of the average size (base diameter) and density of InAs dots for uncapped samples as a function of the InAs coverage determined from atomic force microscopy (AFM). When InAs was increased to 1.71 ML, the average size decreased. On the other hand, the den-

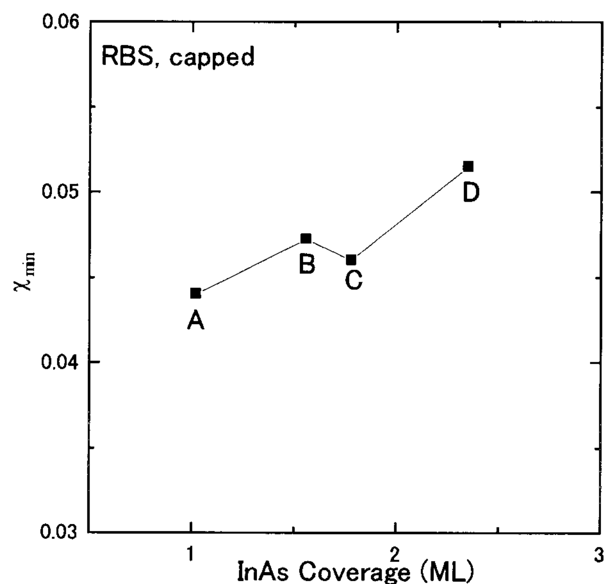

FIG. 3. Summarized $\chi_{\min}$'s of RBS channeling experiments for capped samples.

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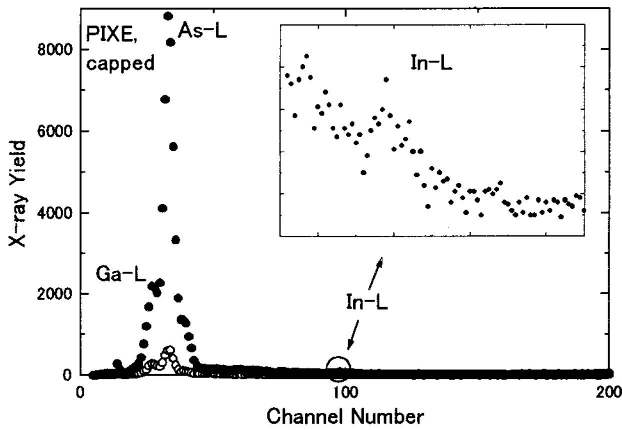

FIG. 4. Typical PIXE spectra for a capped sample. Inset shows a magnified spectrum for In.

sity drastically increased. The average size increased with InAs thickness above 1.8 ML. We note that these trends agree with some reports.[3,6] From Fig. 5, we saw that InAs QDs are largely deformed because χ_{min}'s for In in sample B, C, and D, in which dots are formed, are much larger than that in sample A having no dots. Also from Figs. 5 and 6, we can see that χ_{min} for In positively correlates with the dot size. This means that the deformation of InAs QDs increased with increasing dot size. In this sense, it is comprehensible that χ_{min} for RBS, As, and In in sample C are smaller than those in sample B. On the contrary, the value of χ_{min} for Ga increases when the InAs coverage was changed from 1.56 to 1.78 ML. This suggests that the Ga atoms diffused to form a part of dots.

In order to confirm the diffusion, we performed chemical etching of uncapped samples (sample E, F, G, H, I, and J) with concentrated HCl for 1 min to remove InAs dots and a

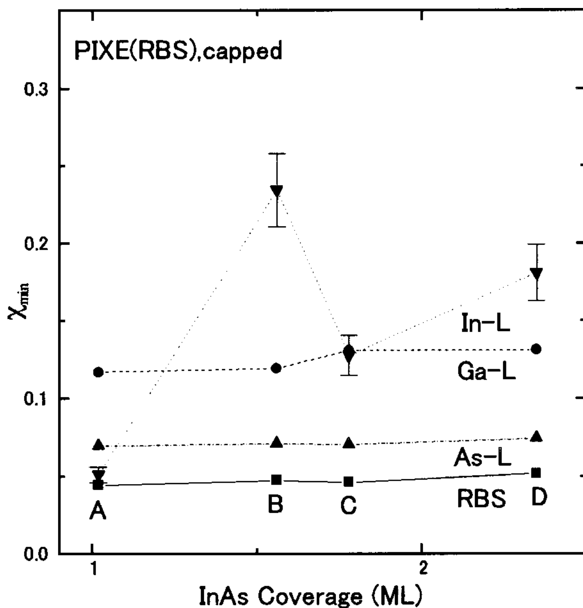

FIG. 5. Variation of the normalized minimum yield for each element and RBS as a function of the InAs coverage. All samples have 150 nm GaAs capping layers.

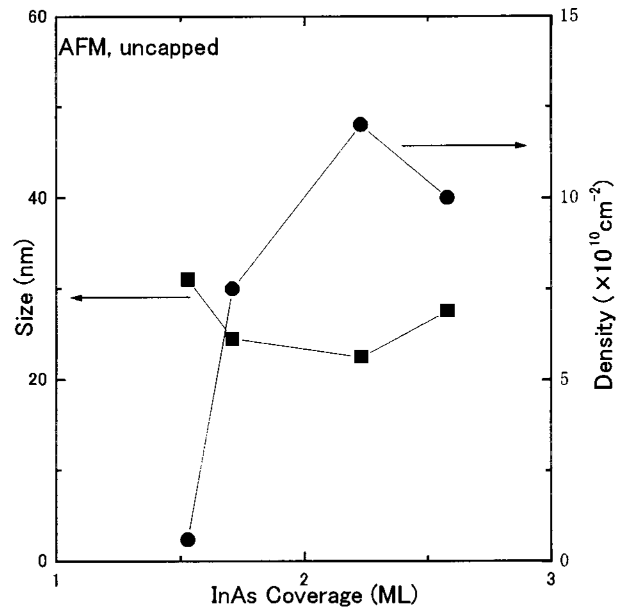

FIG. 6. Variation of the average size (base diameter) and density of the InAs QDs as a function of the InAs coverage for uncapped samples.

wetting layer selectively. After the treatment, we carried out similar RBS experiments with 1.00 MeV He^{+} ions. If the diffusion of Ga atoms exists, In signals are expected to remain, since HCl cannot remove InGaAs.[14]

Figure 7 shows typical RBS channeling spectra for the as-grown uncapped InAs QDs on GaAs(100) (sample F). Channeling effect was not clearly seen due to its native oxidation. Magnified RBS channeling spectra around In signals of these samples are shown in Fig. 8. In Fig. 8, an open circle indicates the spectrum for the etched sample. In Fig. 8(a), we can see the In signals even for the etched samples. This fact and results of AFM observation[14] indicate the existence of the diffusion of Ga atoms into InAs QDs. AFM images of the both as-grown and etched samples are shown in Fig. 9. We recognize the small dots even after etching in Fig. 9(b). However, In signals of the etched samples are not always observed. Indeed, In signals of the etched sample F [see Fig. 8(b)] as well as sample E were hardly observed.

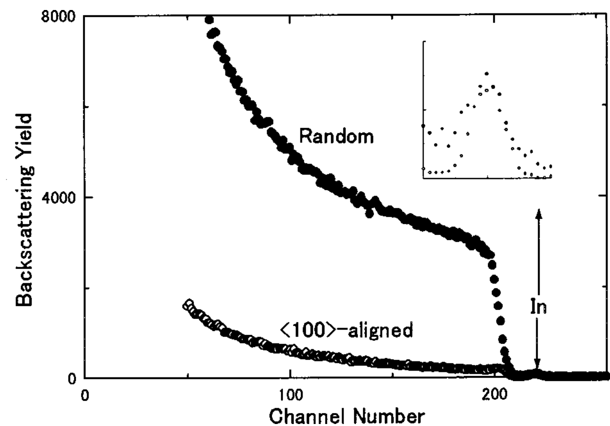

FIG. 7. Typical RBS/channeling spectra for as-grown InAs QDs on GaAs(001) with 1.00 MeV He^{+}. Inset is magnified spectra of the In signals backscattered from InAs QDs and wetting layer.

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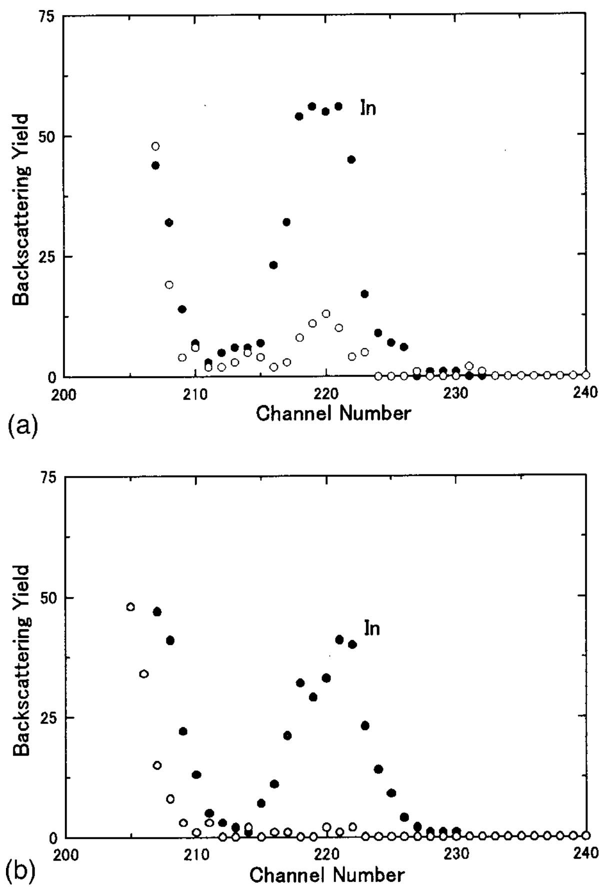

FIG. 8. Magnified RBS spectra of In for as-grown and etched InAs QDs with concentrated HCl for 1 min. (a) sample I (InAs layer thickness: 2.23 ML) and (b): sample F (InAs layer thickness: 1.53 ML).

Figure 10 shows residual In signals of an etched sample normalized by those of the as-grown one as a function of the InAs coverage. In samples E and F [Fig. 8(b)], the residual In signals were very small. However, in sample G, about 12% In signals were observed. The difference in the growth time for InAs is only a couple of seconds. Therefore, the simple diffusion theory does not seem to explain this difference. Rather, this result indicates that there are two 3D growth processes. At ~1.53 ML, InAs QDs grow without any Ga diffusion. On the other hand, InGaAs dots grow when InAs coverage reaches 1.71 ML. Therefore, this suggests that some growth process change occurred in addition to the 2D–3D mode change. In other words, there are three regions of growth suggested. These are 2D growth without Ga diffusion (I), 3D growth with Ga diffusion (II), and 3D growth with Ga diffusion (III) as we increase the InAs coverage.

On the contrary, the correspondence between the capped and uncapped samples is still a subtle issue. To be precise, we have to take those features into account when comparing the capped and the uncapped: (1) change in the deformation of dots when cap layer was overgrown, (2) the deformation of GaAs cap layer especially close to the dot, and (3) additional diffusion between the dots and cap layers. These are issues to be addressed in the future. It should be noted that the three regions specified are for the uncapped samples and are free from the correspondence between the capped and the uncapped. The remarkable thing is that, in spite of these complexities, the strong correlation between deformation of capped samples to the three regions of uncapped samples is easily explained by the diffusion of Ga from the GaAs substrate to the dots.

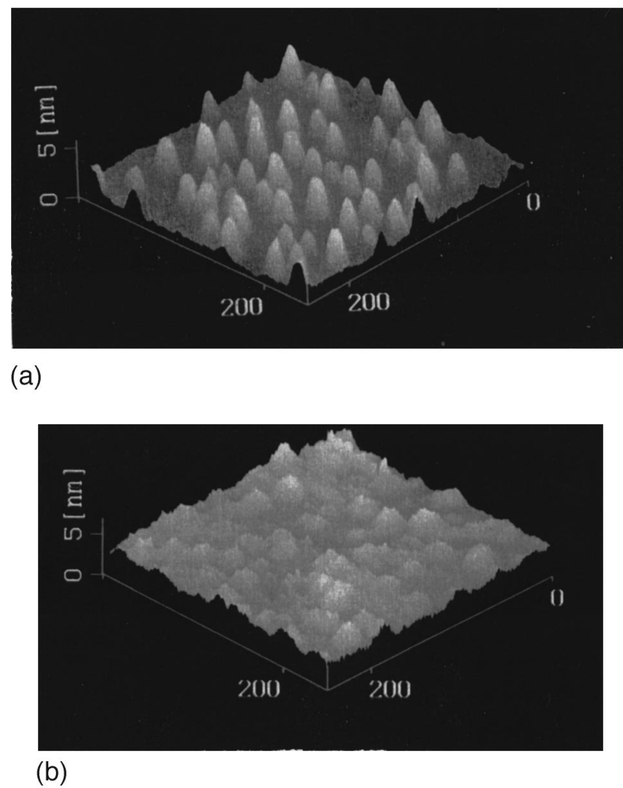

FIG. 9. AFM images of InAs QDs (a) as-grown and (b) etched (concentrated HCl for 1 min) samples.

IV. SUMMARY

In summary, by using ion channeling, we characterized lattice deformation of InAs self-assembled QDs grown by

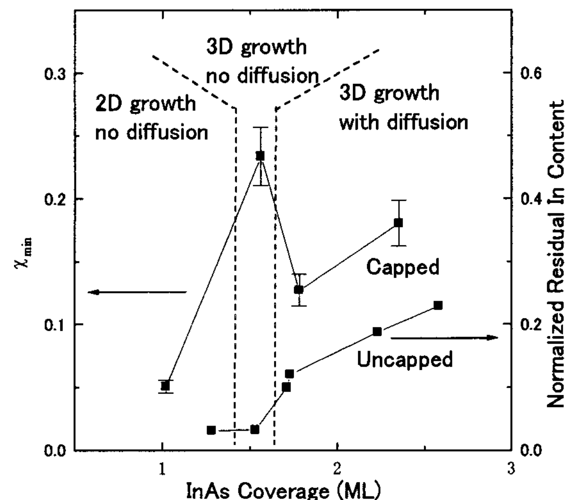

FIG. 10. Residual In content of an etched sample normalized by the In of as-grown sample as a function of the InAs coverage. For comparison, $\chi_{\min}$'s for In are also shown.

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MBE. From the RBS and PIXE experiments, the accurate value of the InAs coverage was obtained even for the capped samples. It was shown that the magnitude of lattice deformation from the GaAs host lattice of the capped samples positively correlated with the dot size. A change in the amount of Ga diffusion into InAs QDs with varying InAs coverage was suggested by using RBS, and confirmed by the etching experiments of the uncapped samples. No Ga atoms were contained in InAs QDs when InAs coverage was 1.53 ML. Thus, diffusion-free 3D growth region and diffusion-enhanced growth region are suggested to exist.

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