Effect of annealing on formation of self-assembled (In,Ga)As quantum wires on GaAs (100) by molecular beam epitaxy

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The role of annealing for (In,Ga)As self-organized quantum wire (QWR) formation on GaAs (100) during growth of (In,Ga)As/GaAs superlattice (SL) structures is studied by x-ray diffraction (XRD), atomic force microscopy (AFM), and photoluminescence (PL) spectroscopy. XRD and AFM evidence that annealing after the supply of each layer of elongated (In,Ga)As quantum dots (QDs) in the SL is the crucial process for QWR formation. We conclude that during annealing, the shape anisotropy of the QDs is enhanced due to anisotropic mass transport and the QDs become connected along the [0-11] direction. Strain reduction by In desorption, revealed by XRD and PL, which accompanies this process, then results in well defined, uniform QWR arrays by repetition in SL growth. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1506191]

I. INTRODUCTION

The formation of self-assembled quantum wires (QWRs) and quantum dots (QDs), based on the Stranski–Krastanov (S–K) growth mode, is extensively investigated with prospect for realization of high-quality dislocation-free nanostructures in strained systems.[1–3] This is motivated by the proposed enhancement of performance of nanostructure active region optoelectronic devices, most prominent, ultralow threshold current semiconductor lasers.[4] The focus of most reports is on QDs, while formation of QWRs could be realized only in very limited material systems, such as InAs/InP (100).[5,6] In the most widely investigated material system, (In,Ga)As/GaAs (100),[1–3] it appeared to be difficult to form well- defined QWRs, although elongated QDs have been observed under specific growth conditions.[7,8] Only recently it has been demonstrated that well- defined (In,Ga)As QWRs with several μm length can be obtained on GaAs (100)[9] by combining elongated island formation at an elevated temperature with a superlattice (SL) growth approach. The detailed mechanism for their formation, however, remains to be clarified. In this article, we identify the role of annealing for formation of these self-organized (In,Ga)As QWRs on GaAs (100). We find that annealing is the crucial process in addition to the SL growth.

II. EXPERIMENTAL DETAILS

The samples were grown on GaAs (100) substrates with miscut smaller than 0.05° by conventional solid-source molecular beam epitaxy (MBE). After growth of a 200 nm thick GaAs buffer layer at 580 °C, the samples were cooled down to 540 °C for the growth of (In,Ga)As. For sample A, without annealing, the $In_{0.36}Ga_{0.64}As$ / GaAs (2.6 nm / 16 nm) SL was grown continuously at 540 °C with growth interruption time of 15 s after each (In,Ga)As layer for stabilization. For the $In_{0.36}Ga_{0.64}As$ / GaAs (2.6 nm / 16 nm) SL structure in sample B, each (In,Ga)As layer was capped with 0.9 nm GaAs at 540 °C without growth interruption, and then annealed at 580 °C for 2 min before GaAs growth was completed. On top of the last GaAs layer of the SL, for both samples, the 2.6 nm $In_{0.36}Ga_{0.64}As$ layer was repeated without annealing. The number of SL periods was 15. The growth rates of GaAs and $In_{0.36}Ga_{0.64}As$ were 0.067 and 0.104 nm/s, respectively, which were calibrated by high-resolution x-ray diffraction (XRD) measurements of (In,Ga)As/GaAs SL structures grown at 480 °C where In desorption is negligible. The As_4 beam equivalent pressure was kept at 1.0×10^{-5} Torr. The structural properties of the samples were characterized by XRD and atomic force microscopy (AFM) in air. For photoluminescence (PL) measurements, the 512 nm line of a Nd–YAG laser was used as excitation source with an excitation power density of 0.2 W/cm^2. The PL was dispersed by a single monochromator and detected by a cooled (In,Ga)As charge-coupled device.

III. RESULTS AND DISCUSSION

Figure 1 shows the XRD spectra in the vicinity of the (311) glancing exit reflection of samples A and B, respectively. In this geometry, XRD is most sensitive to detect the lateral periodicity of modulated structures[9] due to a maximized coherent path and can be evaluated like a multiple-slit Fraunhofer diffraction.[10,11] For sample A, clear satellite peaks or shoulders are observed in addition to the peak from the substrate (plus zeroth order) for incident beam parallel to both the [011] and [0-11] directions, indicating the formation of stacked, laterally ordered QDs. From the spacing between the satellite peaks, the average periodicities along the [011] and [0-11] directions are determined to be 105 and about 130 nm, respectively. The broader peaks or shoulders observed in the [01-1] direction indicate less defined ordering and/or size distribution along this direction, compared to that along [011]. For sample B (with annealing step), the satellite peaks for the x-ray beam along [011] indicate the average period-

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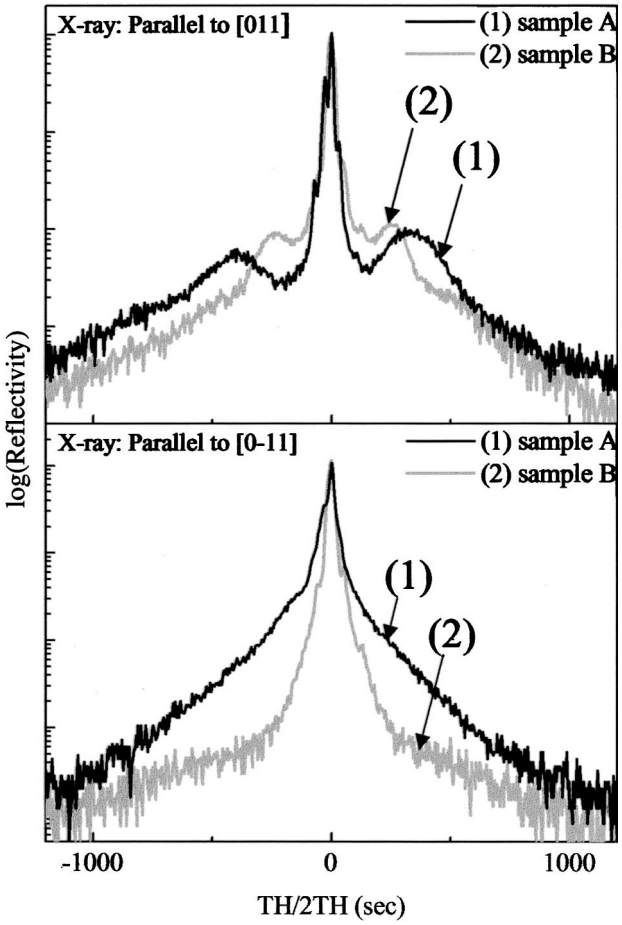

FIG. 1. XRD spectra around the asymmetric (311) glancing exit reflection of samples A and B. The top (bottom) is measured for the x-ray beam parallel to the [011] ([0-11]) direction.

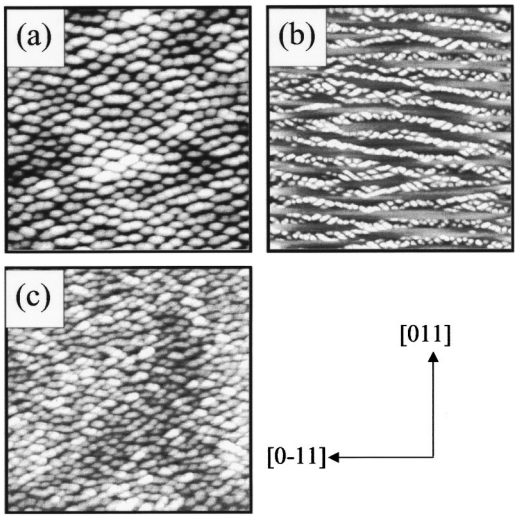

FIG. 2. AFM images of (a) sample A and (b) sample B, respectively. (c) is the AFM image of (In,Ga)As QDs on GaAs (100). The black-to-white height contrast of (a), (b), and (c) is 10, 15, and 10 nm, respectively. The scan field is 2 μm $\times$ 2 μm.

icity along [011] to be 165 nm, which is slightly larger than that for sample A. On the other hand, only the substrate peak is visible with the x-ray beam parallel to the [0-11] direction. This indicates a homogeneous structure along [0-11] and, thus, the transformation from the (In,Ga)As QDs to the uniform QWRs along [0-11] due to the annealing process. The AFM observations confirm the sample structures. Figures 2 (a) and 2(b) depict the AFM images of samples A and B. For reference, the AFM image of 2.6 nm $In_{0.36}Ga_{0.64}As$ grown directly on GaAs (100) under the same conditions is shown in Fig. 2(c). Comparing Figs. 2(a) and 2(c), increased uniformity and a two-dimensional arrangement of QDs is observed after SL growth, which is consistent with previous reports and is attributed to ordering by correlated QD growth due to vertical strain mediation.[12,13] On the other hand, a clear one-dimensional QD array along [0-11] is observed for sample B [Fig. 2(b)]. Taking the positions of the QDs as probes of the lateral strain field minima at the surface, where they preferentially nucleate, this confirms that the underlying (In,Ga)As structure has changed from a two-dimensional QD array to one- dimensional QWR arrays due to the annealing process. The average periodicities in the [011] direction of samples A and B measured from the AFM images are about 110 and 150 nm, which are consistent with the XRD results. Additional information on the formation of QWRs is gained from reflection high-energy electron diffraction (RHEED) inspected during growth. RHEED develops a spotty pattern for each (In,Ga)As layer due to QD formation. The RHEED pattern remains spotty after growth of the 0.9 nm GaAs. The QDs do not completely collapse during capping due to their large size and height for the growth at a high temperature, which has been reported for dots grown at a lower temperature[14] and is consistent with the stability of large dots formed at low growth rate.[15] During the annealing process, the RHEED pattern then changes from spotty to rather streaky, indicating significant change of the structure. In recent postgrowth annealing experiments of buried InAs QDs in GaAs, an increased size and decreased In composition were found.[16–18] Finally, the QDs may change into two-dimensional quantum wells.[17] Annealing temperatures, however, were significantly higher. In contrast, for our structure, the GaAs capping layer is that thin so to allow significant mass transport at the surface to change the QD structure. As indicated by RHEED, the surface flattens which is known to result in moundlike structures with pronounced elongation along [01-1] due to anisotropic adatom surface migration.[19] From these results, we propose the following mechanism for QWR formation:

(1) (In,Ga)As QDs form. Due to growth at high temperature, the QDs are elongated along [0-11].
(2) During annealing, elongation of the (In,Ga)As QDs is enhanced due to anisotropic migration.
(3) The QDs become connected along the [0-11] direction, forming QWRs.
(4) In the subsequent (In,Ga)As layer, the QDs form on the GaAs surface above the QWR regions due to strain mediation.
(5) During annealing, the (In,Ga)As QDs elongate along [0-11].

By repeating these processes in SL growth, well-defined QWRs are formed. Vice versa, it is inherent from sample A

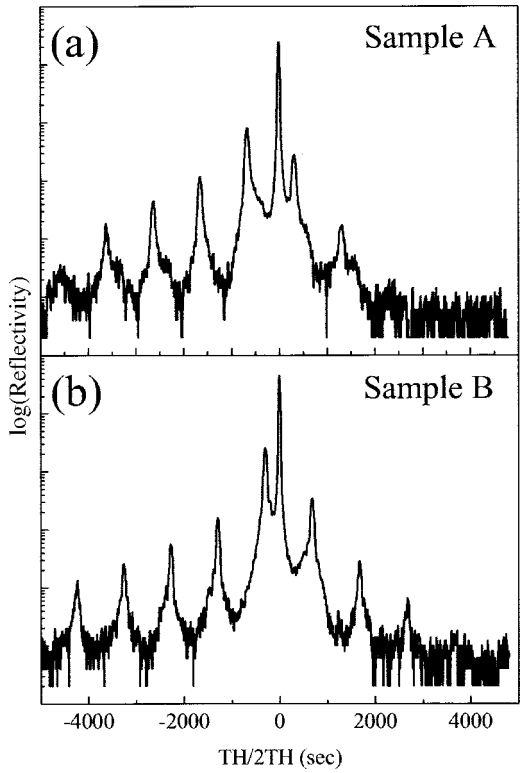

FIG. 3. XRD spectra around the symmetric (400) reflection of (a) sample A and (b) sample B.

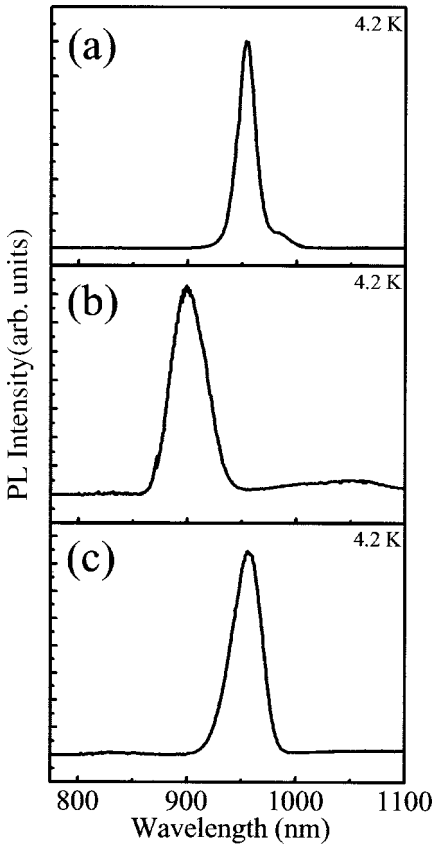

FIG. 4. Low temperature PL spectra of (a) sample A and (b) sample B. (c) shows the PL spectrum of capped (In,Ga)As QDs grown on GaAs (100).

that the maintenance of a QD structure without an additional elongation process is not leading to the purely one-dimensional QWR structure in sample B confirmed from XRD and the (In,Ga)As QD positions in the top layer. For more than 10 SL periods, the top layer indicates that the length of the QWRs easily exceeds several μm and the QWR SL serves as a template for natural one-dimensional QD ordering.[20]

For QWR formation versus QDs, it is, moreover, suggested that the total strain energy needs to be reduced to guarantee uniform connection of the QDs because elastic strain relaxation along the wire direction can not occur. Indeed, in our structure, an additional effect of annealing is strain reduction by In desorption. This is confirmed by XRD measured in the vicinity of the symmetric (400) reflection for samples A and B shown in Figs. 3(a) and 3(b). The absence of dislocations has been confirmed by comparison with the asymmetric (422) reflection. Table I shows the results of average In composition and SL periodicity for sample A and B in comparison to the nominal values determined from the SL grown at 480 °C. For the evaluation of XRD, two- dimensional layer structures have been assumed (therefore, the measured In composition in samples A and B might be slightly underestimated due to lateral elastic strain modulation). Even in the case of sample A, without an annealing step, the average In composition is markedly reduced from 0.050 to 0.036, suggesting the onset of In desorption due to the high growth temperature of 540 °C. In the case of sample B, with annealing, the average In composition is drastically reduced to 0.016. Almost 60% of In is desorbed from the surface during growth and annealing. The difference of the SL periodicities is attributed to run-to-run growth rate variations of about 3%, which can not account for the measured drop of In composition. It is interesting to note, that the In composition of sample B, if originally supplied, is too low to form a three-dimensional (In,Ga)As structure. The (In,Ga)As layer would remain flat. Hence, we conclude that QWR formation requires initial formation of QDs during deposition of (In,Ga)As with sufficiently high In composition, commonly observed in the S–K growth mode. The following annealing process is then necessary to enhance the QD elongation by anisotropic surface migration and simultaneously reduce the In composition—most probably below the critical value for QD formation—to produce the uniform (In,Ga)As QWRs on GaAs (100). The lower In composition in sample B also leads to the larger lateral periodicity along [011] compared to that in sample A.

TABLE I. Average In compositions and SL periodicities for samples A and B, in comparison to the nominal value determined from XRD.

	Average In composition in SL	Periodicity of SL
Nominal supply	0.050	18.6 nm
Sample A	0.036	19.1 nm
Sample B	0.016	19.2 nm

The low-temperature PL spectra from samples A and B are shown in Figs. 4(a) and 4(b), respectively. For compari-

son, the PL spectrum of a single-capped $In_{0.36}Ga_{0.64}As$ (2.6 nm) QD layer grown at 540 °C is shown in Fig. 4(c). In sample A, a narrow peak is observed at 954 nm (1.30 eV). Compared to Fig. 4(c), the linewidth is reduced from 37 to 23 meV due to QD stacking while the peak energy is almost unchanged. This agrees with the improved size uniformity due to stacking,[12] observed in AFM. For sample B, a broader peak with 47 meV linewidth is observed at 901 nm (1.38 eV). The large blueshift from 1.30 to 1.38 eV is consistent with the In desorption and QWR formation. The larger linewidth might originate from residual size and composition fluctuations introduced during annealing and the reduced height of the QWRs. Surface mass transport and In desorption involved in QWR formation clearly distinguish this process from postgrowth annealing of buried (In,Ga)As QDs in a GaAs matrix,[16–18] where bulk diffusion and intermixing, and conservation of total In composition have not been observed to lead to formation of QWRs.

IV. CONCLUSION

We have studied the role of annealing for (In,Ga)As self-organized QWR formation on GaAs (100) during growth of (In,Ga)As/GaAs SL structures by MBE. The formation of QWR requires (1) initial formation of QDs by deposition of (In,Ga)As with sufficiently high In composition and (2) annealing. During the annealing process, the QD elongation is enhanced by anisotropic surface migration and the QDs become connected. In addition, strain reduction by In desorption during annealing is necessary to realize the QWRs of several μm length, which is revealed by XRD and PL.

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