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Self-Formation of Uniform InAs Quantum Dots and Quantum-Dot Chains

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Uniform InAs quantum dots (QDs) were demonstrated on GaAs(001) substrates by molecular beam epitaxy (MBE) via Stranski-Krastanov (SK) growth mode. Enhanced surface migration of growth species due to low growth rate and low arsenic pressure played an important role for reducing a size fluctuation of SK QDs. Growth conditions of a GaAs capping layer also influenced the inhomogeneous broadening in the QD energy level. We successfully obtained the narrowest photoluminescence linewidth of 17 meV for a single InAs-QD layer. One-dimensional self-alignment of InAs QDs (QD chain) was also fabricated on GaAs(001) substrates only by MBE growth without lithography and etching processes. InAs QD chains were selectively formed on self-formed GaAs mesa-stripes, grown on strained InGaAs/GaAs buffer layer.

Key words: quantum dots, InAs, quantum-dot chain, molecular beam epitaxy, GaAs

1. INTRODUCTION

Self-formation techniques based on Stranski-Krastanov (SK) growth mode are much attractive for a fabrication of dense quantum dots (QDs) with high crystal quality. The SK growth technique has been studied actively for more than ten years [1]. However, the formation mechanism of SK QDs has not been understood clearly. In order to realize some device applications [2], control of SK QDs must be more improved and is still an open challenge. In particular, a large inhomogeneous broadening in QD energy level is an important problem. So far, several growth techniques have been attempted to fabricate uniform QDs [3-5], however, it is difficult to achieve a narrow photoluminescence (PL) linewidth of less than 20 meV for a single QD layer. Recently, we have demonstrated a highly uniform single-QD-layer of the InAs, which revealed 18.6 meV in PL linewidth [6]. In addition, a precise control of dot positions and a selective arrangement of QDs are also needed for some device applications. Lithography and etching techniques are often used for a selective fabrication of QDs on non-planar substrates [7]. We have been investigating one-dimensional (1D) self-alignment of InAs QDs (QD chains) on a strained buffer layer without lithography and etching processes [8]. Recently, InAs QD chains were successfully formed on self-formed GaAs mesa-stripes, grown on the strained InGaAs/GaAs buffer layer [9].

In this paper, first we present self-formation of highly uniform InAs QDs by molecular beam epitaxy (MBE) growth via the SK mode. Control of the SK growth and the capping growth are extremely important to a fabrication of uniform QDs. Their growth conditions for the self-formation of uniform InAs QDs are presented, and their physical origin are discussed. Second we present a self-formation technique of 1D alignment of InAs QD chains on the strained GaAs/InGaAs buffer layer.

2. EXPERIMENT

Samples in this study were prepared on GaAs(001) substrates by conventional solid-source MBE. The substrate temperature was calibrated by using the Ga-oxide desorption temperature of 580 ℃ as a reference. Prior to the InAs growth, a 200-nm-thick GaAs buffer layer was grown at 590 ℃. The substrate temperature was cooled down to 500 ℃, and then self-assembled dots of the InAs were formed by using SK growth mode. The InAs coverage was changed from 1.2 to 3.0 monolayers (ML). The arsenic beam equivalent pressure was changed from 3×10^{-7} Torr to 6×10^{-7} Torr. The InAs coverage (1.6-3.0 ML) and the growth rate (0.035-0.17 ML/s) were calibrated based on the critical thickness of the SK-growth mode transition from 2D to 3D by using reflection high-energy electron diffraction (RHEED). The samples grown for PL measurements were capped with a 100-nm-thick GaAs layer.

For PL studies, Ar^+ laser was used for excitation (25 mW), and the luminescence was detected using a cooled InGaAs photodiode. The dot structure was evaluated by using an atomic force microscopy (AFM) and a scanning transmission electron microscopy (STEM) with high-angle anular dark field (HAADF) mode.

The MBE growth condition and the sample structure of InAs QD chains are described at section 3-2.

3. RESULTS AND DISCUSSION

3.1 Uniform InAs QDs

The SK growth process of the InAs on the GaAs substrate is described first. Figure 1 shows a typical relationship between the lateral size and the height of InAs islands, grown at a growth rate of 0.16 ML/s and an arsenic pressure of 6×10^{-7} Torr. The InAs coverage was changed from 1.6 to 3.0 ML. As the growth proceeds until a 2D-3D transition of the growth mode, the height of 2D islands is almost kept at 1-3 ML and the

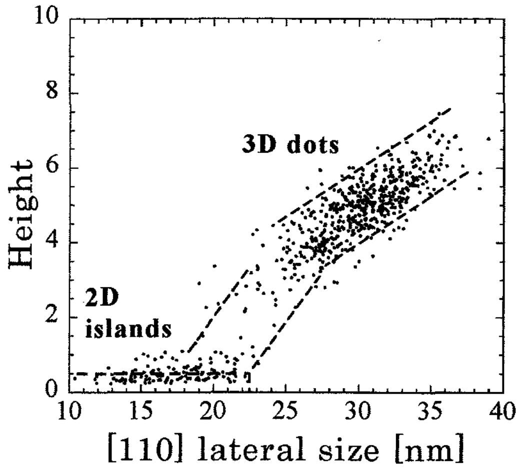

Fig. 1. Relationship between the lateral size and the height of 2D and 3D islands of the InAs (1.6-3.0 ML in coverage), grown on GaAs(001) substrates.

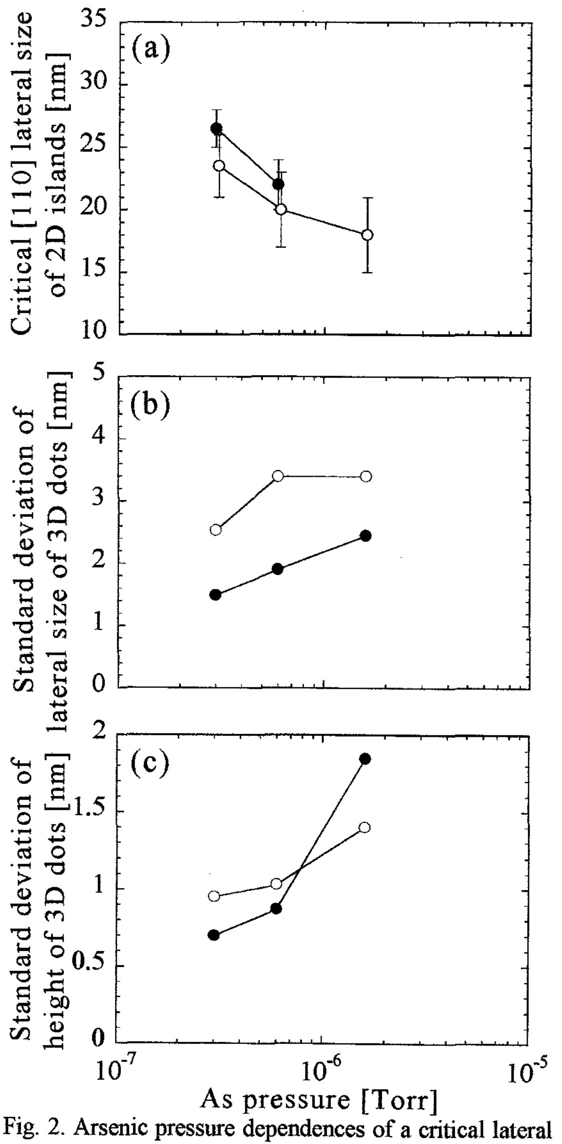

Fig. 2. Arsenic pressure dependences of a critical lateral size of 2D islands (a) and standard deviation of 3D dots (b). InAs growth rates were 0.035 ML (●) and 0.16 ML/s (○), respectively.

lateral size increases. When the lateral size reaches around 20 nm (critical lateral size), the height rapidly increases: the growth mode transits from 2D to 3D because of elastic strain. Here, it should note that inhomogeneous broadening in the critical lateral size of 2D islands provides size fluctuation of 3D dots.

Figure 2(a) shows the growth condition dependence of the critical lateral size of 2D islands. As the growth rate and the arsenic pressure decreases, 2D islands enlarge in the critical lateral size. In case of high growth rate and high arsenic pressure, many multi-stacked 2D layers have been observed by AFM, as reported previously [10]. These results mean that the multi-nucleation mode easily induces the 2D-3D transition on smaller 2D islands (i.e. smaller critical lateral size). This growth situation can be explained by consideration based on the energetic stability of the 3D islanding: the transition energy on multi-stacked 2D islands is lower than that on large single islands. In addition, the multi-nucleation mode will provide various 3D nucleation sites on the 2D islands as compared with the single nucleation-mode. Indeed, size fluctuation in the critical lateral size of 2D islands increases for high growth rate and high arsenic pressure conditions (see a bar of data in Fig. 2(a)). As a result, the standard deviation of 3D dots also becomes large for high growth rate and high arsenic pressure conditions, as shown in Figs. 2(b) and 2(c). Therefore, low growth rate and low

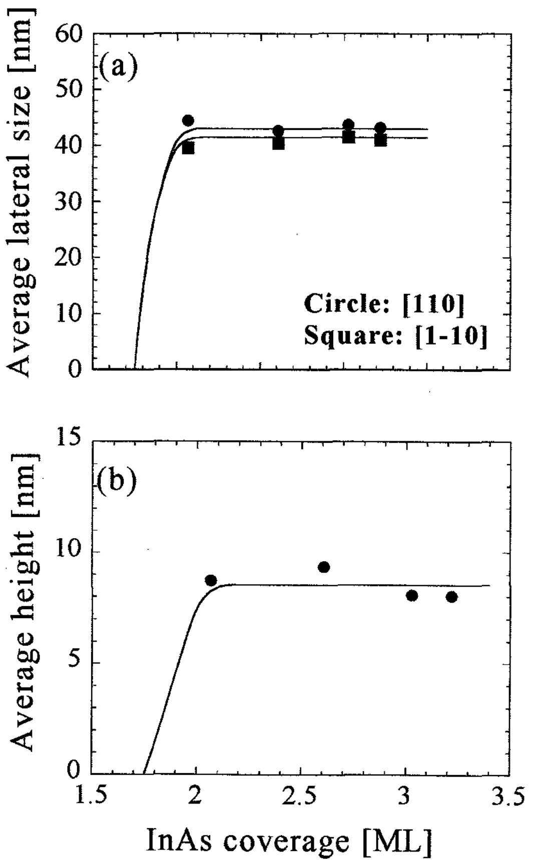

Fig. 3. Average lateral size (a) and average height (b) of InAs dots as a function of InAs coverage.

arsenic pressure are effective growth conditions to achieve uniform 3D nucleation.

The next important point for fabrication of uniform InAs QDs is a precise control of the 3D dot structure. Recently, we have observed a self size-limiting phenomenon of 3D InAs dots, which plays an important role for reduction of the size fluctuation [6,11]. Figure 3 shows the average lateral size (a) and average height (b) of InAs 3D dots as a function of the InAs coverage. The InAs growth conditions were low growth rate (0.035 ML/s) and low arsenic pressure (3×10^{-7} Torr). After the rapid transition from 2D to 3D growth mode, the growth of 3D dots suddenly stops. As shown in Fig. 3, the lateral size and height almost saturate. Also, the dot density is kept constant at about 3×10^{10} cm^{-2}. Therefore, as the growth proceeds, size-limited InAs dots are accumulated, and the size fluctuation becomes narrow. In fact, the PL linewidth decreased with increasing the InAs coverage.

From analyses of the dot structure by using RHEED, AFM and TEM, it was found that the growth stop is related to the facet formation on the side wall of the dot [12]. In short, incorporation of indium adatoms is effectively suppressed on the stable facet. As a result, the size-limiting process accumulates the surface concentration of indium adatoms. Indeed, the RHEED pattern revealed the indium-stabilized surface, which also induced coalescence of size-limited dots [12].

Figure 4(a) shows a typical AFM image of the size-limited InAs dot covered by {101} facets ({111}B facets also appear partially on the side wall). The crystal orientation of the facet strongly depends on the growth conditions (growth rate and arsenic pressure), as shown in Fig. 4(b). When the growth rate and arsenic pressure decrease, the facet plane transits from {136} to {101} plane. From a viewpoint of reducing the size fluctuation, high aspect ratio is desirable structure. Therefore, low growth rate and low arsenic pressure conditions ({101} facet formation) are further expected for suppression of

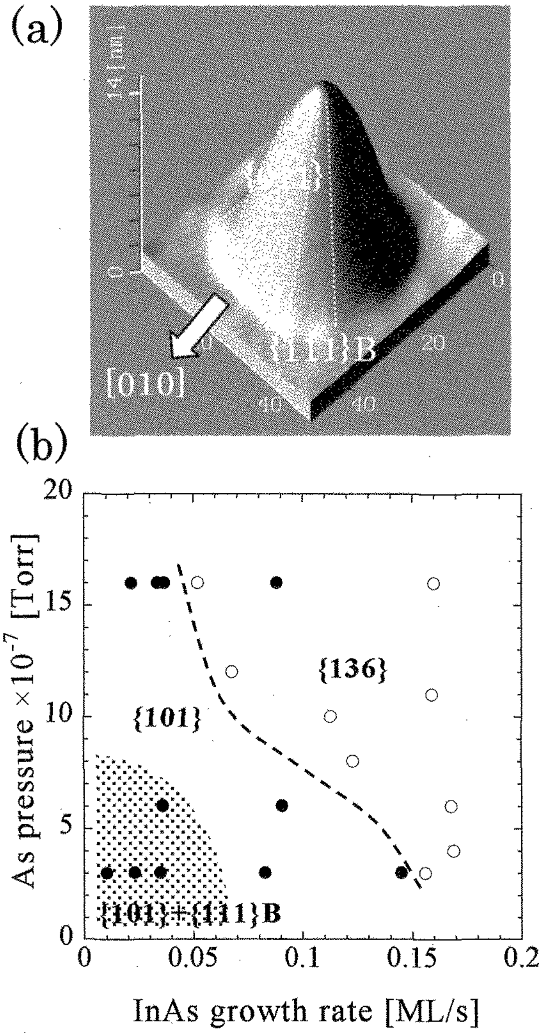

Fig. 4. AFM image of the InAs dot, which is mainly covered by (110) facets (a). Relationship between a crystal orientation of the facet and growth conditions (InAs growth rate and arsenic pressure) (b).

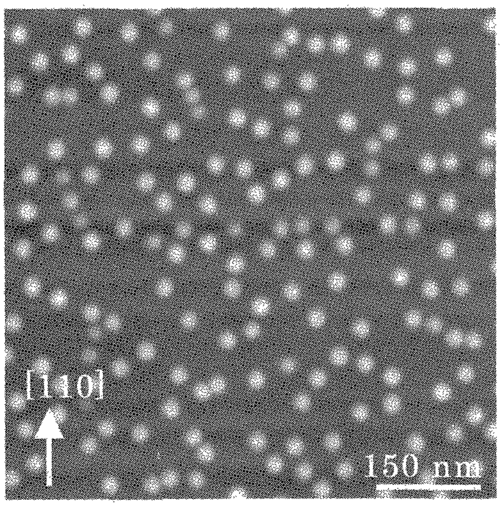

Fig. 5. AFM image of uniform InAs dots, grown at the growth rate of 0.035 ML/s and arsenic pressure of 3×10^{-7} Torr.

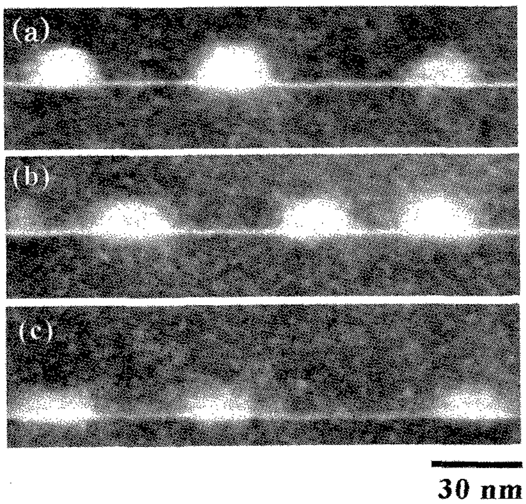

Fig. 6. (110) cross-sectional HAADF-STEM images of InAs QDs embedded by GaAs layer. The growth temperature of GaAs capping layers were 400 ℃ (a), 450 ℃ (b) and 500 ℃ (c), respectively.

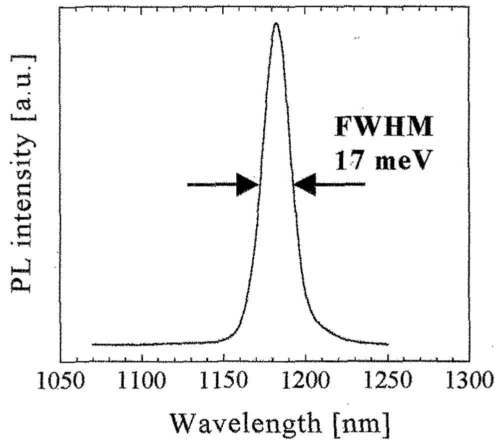

Fig. 7. PL spectrum of uniform InAs QDs. PL temperature was 12 K.

inhomogeneous broadening in the QD size.

Based on those results, we fabricated uniform InAs SK dots by low growth rate (0.035 ML/s) and low arsenic pressure (3×10^{-7} Torr). Figure 5 shows an AFM image of uniform InAs dots. As mentioned before, the dot is mainly covered by {101} facets. The standard deviations are 1.6 nm (4 %) for the lateral size and 0.5 nm (8 %) for the height, which are the narrowest size-fluctuation than that of conventional SK dots.

The next growth process, a GaAs capping growth on uniform InAs SK dots, is also a noteworthy process for realization of uniform InAs QDs because the capping growth frequently modifies the dot structure. In particular, the substrate temperature strongly influences on the dot shape. Figure 6 shows (110) cross-sectional HAADF-STEM images of embedded InAs QDs as a function of the substrate temperature of the GaAs capping layer. In general, a low temperature growth of the GaAs provides a low crystal quality. However, as the substrate temperature increases, the SK dot structure remarkably changes. After the capping growth at 500 ℃, the dot height decreased from 11 nm to about 6 nm, and the lateral size also decreased from 28 nm to about 24 nm. Such modification of the dot structure is mainly attributed to the indium surface segregation and the indium desorption during the capping growth and the growth interruption. To keep a high aspect ratio of InAs QDs and a high crystal quality, the GaAs capping growth at 450 ℃ was done under low growth rate (0.14 nm/s) and low arsenic pressure (6×10^{-7} Torr) conditions. In this capping growth, the average dot height was maintained at about 8 nm and the average lateral size was about 24 nm.

Figure 7 shows a low-temperature (12 K) PL spectrum of uniform InAs QDs with the GaAs capping layer, grown at above selected conditions. The peak wavelength is 1180 nm, and the strong PL intensity was kept until about 200 K. The PL linewidth is 17 meV, which is the narrowest value as compared with ever reported for the single QD layer with a high dot density ($> 10^{10}$ cm^{-2}).

3.2 InAs QD Chains

SK dots are preferentially formed near step edges because of the strain relaxation. Thereby, if the straight step structures can be prepared on the surface, it is possible to achieve 1D alignment of SK dots along the step edge. However, control of the fine step structure is difficult, and high selectivity in the self-formation on the step edge is needed to suppress the normal random deposition on the other area. Here we present a self-formation technique of 1D alignment of InAs QDs (QD chains) on the strained GaAs/InGaAs buffer layer.

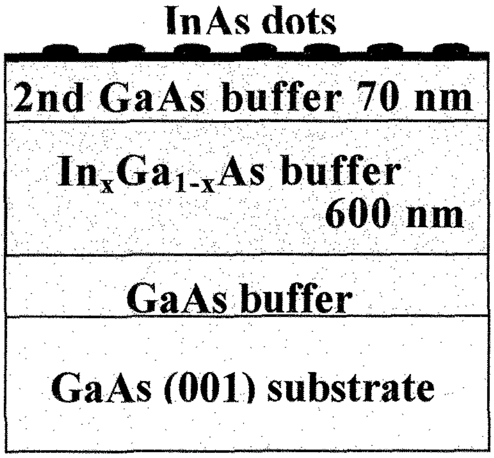

Fig. 8. Schematic diagram of sample structure for a fabrication of InAs QD chains.

Figure 8 shows a schematic diagram of the sample, prepared by the MBE growth. The GaAs buffer layer was grown at a substrate temperature of 590 ℃ on GaAs(001) substrates, and then a 600-nm-thick $In_xGa_{1-x}As$ buffer layer (x: 0.21-0.23) was grown at 500 ℃. Additionally, the second GaAs buffer layer with 70 nm in thickness was grown on the corrugated InGaAs buffer layer. Self-assembld InAs QDs were grown on the buffer layer by low growth rate and low arsenic pressure conditions, using same conditions with those of uniform QDs, mentioned at section 3-1. The misfit dislocations were nucleated at the lower and upper InGaAs/GaAs heterointerfaces because the thickness reached the equilibrium critical thickness. We note that misfit dislocations play an important role for the fabrication of InAs QD chains as follows.

Figure 9 shows a SEM image of InAs QDs grown on the GaAs/InGaAs hetero-buffer layer. One can see GaAs strip-shaped patterns and InAs QD chains along the [1-10] direction. The QD chains with a long distance (several μm) mainly locate on the top of the GaAs strip pattern and on the groove between stripes. Sometimes the random deposition of QDs is observed on the central area of the wide GaAs mesa strip. According to our previous report [8], such 1D selective alignment of InAs QDs was due to the misfit dislocations at the GaAs/InGaAs heterointerface. In this structure, the self-formation of long QD chains is related to not only misfit dislocations but also mesa structures.

Figure 10 shows a cross-sectional HAADF-STEM image of InAs QDs grown on the GaAs/InGaAs buffer layer. It is found that the {211}A facet appears on the side wall of the GaAs mesa strip and that the stacking fault generates from the upper heterointerface to the groove between the mesa stripe. That is, the self-formation of the GaAs mesa structure is caused by

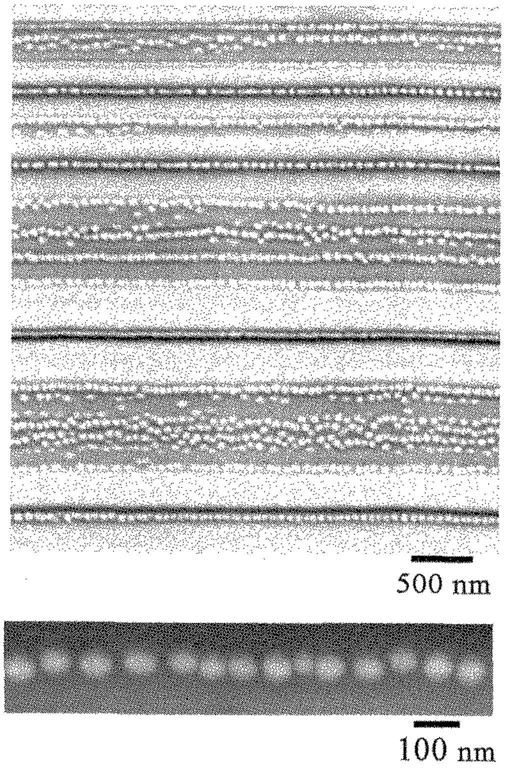

Fig. 9. SEM image of InAs QD chains.

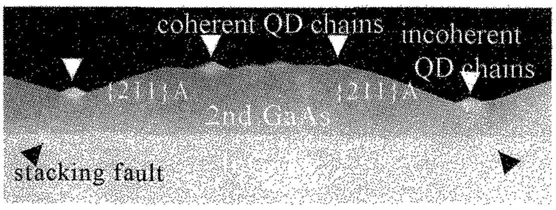

Fig.10. (1-10) cross-sectional HAADF-STEM image of InAs QDs grown on GaAs mesa stripe.

the generation of the stacking fault, which probably induces the local growth pinning of the GaAs buffer layer. When the indium composition of the InGaAs buffer layer was lower than about 0.2, there was no stacking fault at the heterointerface. In this case, the mesa strip was not formed, but the surface corrugation appeared. In Fig. 10, incoherent InAs QD chains due to stacking faults are observed at the groove. However, it is also found that coherent QD chains are formed along misfit dislocations, as mentioned before. These coherent QD chains have high straightness over a long distance as compared with that formed on the GaAs corrugated surface (for example, in case of the GaAs/$In_{0.17}Ga_{0.83}As$ buffer layer). Therefore, the straight mesa-strip structure having the flat {211}A facet induces straight step structures or straight strain-relaxed region along the [1-10] direction. As a result, the QD chain with high straightness is automatically formed on the mesa strip. The presented control technique of the strain and dislocations into the underlying layers is a useful for the selective arrangement of quantum nanostructures without the use of lithographic patterns.

4. CONCLUSION

Uniform InAs QDs were demonstrated by MBE using SK growth mode. In the SK growth of the InAs QDs, the low growth rate and the low arsenic pressure were effective conditions to suppress inhomogeneous broadening in the dot size and shape. These growth conditions enhanced a surface migration of growth species and suppressed multi-nucleation mode of 2D InAs islands, which induced uniform and large 2D critical islands just before the 3D islanding. During the 3D growth, self size-limiting effect was caused by facet formation on the side wall. And then uniform 3D dots were accumulated. Furthermore, the growth condition of the GaAs capping layer was investigated, and, finally we successfully obtained the narrowest PL linewidth of 17 meV.

A new fabrication method of QD chains using a strained GaAs/InGaAs buffer layer was presented. The GaAs mesa-strip pattern was automatically formed on the InGaAs buffer layer without lithography and etching techniques. Coherent QD chains of the InAs were formed on the GaAs mesa strip. The selective self-formation of QD chains was mainly explained by generation of misfit dislocations at the GaAs/InGaAs heterointerface.

5. ACKNOWLEDGEMENT

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6. REFERENCE

[1] Tabuchi, S.Noda and A.Sasaki, Sci. & Tech. of Mesoscopic Structures, eds. S.Namba, C.Hamaguchi and A.Ando, 379, Springer Verlag, Tokyo (1992).
[2] Y.Arakawa and H.Sakaki, Appl. Phys. Lett., 40 (1982) 939.
[3] K. Nishi, H. Saito, S. Sugou and J-S. Lee, Appl. Phys. Lett., 74 (1999) 1111.
[4] I. Mukhametzhanov, Z. Wei, R. Heitz and A. Madhukar, Appl. Phys. Lett., 75 (1999) 85.
[5] R.Murray, D.Childs, S.Malik, P.Siverns, C.Roberts, J-M.Hartmann and P.Stavrinou, Jpn. J. Appl. Phys., 38 (1999) 528.
[6] K.Yamaguchi, K.Yujobo and T.Kaizu, Jpn. J. Appl. Phys., 39 (2000) L1245.
[7] D.S.L.Mui, D.Leonard, L.A.Coldren and P.M.Petroff, Appl. Phys. Lett., 66 (1995) 1620.
[8] K.Yamaguchi, K.Kawaguchi and T.Kanto, Jpn. J. Appl. Phys., 41 (2002) L996.
[9] T.Kanto and K.Yamaguchi, 2nd Int. Conference on Semiconductor Quantum Dots, Tokyo (2002) D-2, p.24.
[10] S.Iwasaki and K.Yamaguchi, Appl. Surf. Sci., 216 (2002) 407.
[11] T.Kaizu and K.Yamaguchi, Jpn. J. Appl. Phys., 40 (2001) 1885.
[12] T.Kaizu and K.Yamaguchi, Jpn. J. Appl. Phys., 42 (2002) 4166.

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