Temperature-Dependent Site Control of InAs/GaAs (001) Quantum Dots Using a Scanning Tunneling Microscopy Tip During Growth

Takashi Toujyou · Shiro Tsukamoto

Abstract  Site-controlled InAs nano dots were successfully fabricated by a STMBE system (in situ scanning tunneling microscopy during molecular beam epitaxy growth) at substrate temperatures from 50 to 430°C. After 1.5 ML of the InAs wetting layer (WL) growth by ordinal Stranski–Krastanov dot fabrication procedures, we applied voltage at particular sites on the InAs WL, creating the site where In atoms, which were migrating on the WL, favored to congregate. At 240°C, InAs nano dots (width: 20–40 nm, height: 1.5–2.0 nm) were fabricated. At 430°C, InAs nano dots (width: 16–20 nm, height: 0.75–1.5 nm) were also fabricated. However, these dots were remained at least 40 s and collapsed less than 1000 s. Then, we fabricated InAs nano dots (width: 24–150 nm, height: 2.8–28 nm) at 300°C under In and As4 irradiations. These were not collapsed and considered to high crystalline dots.

Keywords  Quantum dot · Site control · In situ · Scanning tunneling microscopy · Molecular beam epitaxy

Introduction

Recently, studies on the semiconductor self-assembled quantum dot (QD) have great attentions because of their applications in optoelectronics, such as high-efficiency QD lasers, QD solar cells (QDSCs), etc. Especially, QDSCs [1] are easily turned to the sunlight spectrum, since it can select the photo-absorption wave length by controlling QD size and superlattice structures to form an intermediate band or a miniband rather than a multiplicity of discrete quantized levels [2]. These QDs must be uniform in size and periodically distributed in all three dimensions (3D) to achieve the predicted high conversion efficiencies [3]. The most popular fabrication technique of QDs is to take advantage of spontaneous self-assembly or self-organization mechanism of coherent 3D islanding during growth known as a Stranski–Krastanov (S–K) growth in lattice-mismatched epitaxy. However, it was governed by statistics, so that QDs nucleate more or less randomly on semiconductor surface. This random nucleation makes it difficult to address each individual self-assembled QD, then several groups reported site-selective QD growth methods: nano-jet probe method [4], applying voltage from an STM tip [5], and growth on patterned substrates [6, 7]. However, taking out a sample from a growth chamber, or quenching a substrate temperature and stopping As4 supply during the substrate processing stage of these methods, causes the crystalline deterioration of the QDs. To overcome this problem, operating a QDs arrangement during MBE growth by a STMBE system that placed a scanning tunneling microscope (STM) inside a molecular beam epitaxy (MBE) growth chamber and performs true in situ imaging during the MBE growth [8] was efficacious. Using this system, Tsukamoto et al. reported in situ observation of an InAs wetting layer (WL) and QDs formation on GaAs (001) at 430°C [9]. In their report, tiny alloy fluctuations in the WLs, such as atomistic point defects (Ga-rich sites), were important in controlling QD nucleations. Migrating In atoms tends to distribute the Ga-rich sites at the InAs WL. Same phenomenon appeared at 500°C [10]. Therefore, in this paper, using this phenomenon, we tried to fabricate high crystalline InAs nano dots by applying voltage from an STM tip at different temperatures without breaking MBE growth environments.
Experimental Details

All experiments were carried out with the STMBE system. We used a GaAs (001) just substrate. All samples were prepared at same procedure: after removing oxides at 580°C, about 170 nm of a GaAs buffer layer was grown at 560°C under Ga and As4 fluxes, 1.0 × 10⁻⁵ Pa and 6.0 × 10⁻³ Pa, respectively. After the GaAs buffer layer growth, about 1.5 monolayers (ML) of an InAs WL were grown at 500°C under In and As4 fluxes, 1.28 × 10⁻⁶ Pa and 6.0 × 10⁻⁴ Pa, respectively. After forming the InAs WL, a substrate temperature was decreased from 500°C to that of described in each observation. In the observation at 430°C, it was decreased from 500 to 430°C under As4 irradiation. When the substrate temperature was stabilized, STM units were combined and immediately started in situ STM observation. In our observations, we investigated the best STM parameters (a tip bias, a tunneling current, and a scan speed) for each case. During the observation, an STM tip scanned from left to right and moved from bottom to top of the image. After a thermal drift reducing, using the STM tip, we applied voltage ‘from a reverse bias to a forward bias’. The time of applying voltage was 0.052 s, and a voltage value was increased every 0.00026 s by a step function, and then observed the surface structure transitions.

Results and Discussion

Surface Structure Transition at 50–150°C Without As4

At first, we confirmed the surface structure transition by applying voltage at low temperatures. After the InAs WL growth, we applied voltage on the InAs WL without As4. Figure 1a–c was the STM images of surface structure before applying voltage at 50 and 150°C. White points were indicating the point of applying voltage. During in situ STM observation (a tip bias was +1.0 V, a tunneling current was 0.4 nA, and the scan speed was 1,500 nm/s), we applied voltage from −6 to +6 V at 50°C. A surface structure transition was shown in Fig. 1(a’). In this image, any surface structure transition was not confirmed. Then, similar observation was operated at 150°C. A tip bias was +1.0 V, a tunneling current was 0.2 nA, and the scan speed was 2,500 nm/s. We applied voltage from −6 to +6 V. The surface structure transition was shown in Fig. 1(b’), a hole structure (depth: 4 nm, width: 20 nm) was appeared at the point of applying voltage, and a ring structure (93 × 101 nm²) was appeared around the hole structure. This hole structure dug the WL and reached to the GaAs substrate. Next, we applied voltage from −3 to +3 V at 150°C as shown in Fig. 1c. A tip bias was +1.0 V, a tunneling current was 0.2 nA, and the scan speed was 1,500 nm/s. A hole structure (depth: 1.4 nm, width: 15 nm) was appeared at the point of applying voltage, and a ring structure (45 × 50 nm²) appeared around the hole structure as shown in Fig. 1(c’). This ring structure was smaller than that of Fig. 1(b’). This result indicated that the size of the surface structure transition can be controlled by the amount of voltage, and it needs at least 150°C.

Surface Structure Transition at 240°C Without As4

After the InAs WL growth at 500°C, a substrate temperature has decreased to 240°C and stopped As4 supply. Figure 2 shows the continuous STM images of the surface structure transitions by applying voltage from −0.5 to +0.5 V. A tip bias was −0.1 V, a tunneling current was 0.2 nA, and the scan speed was 5,000 nm/s. A dot structure (width: 38 nm, height: 1.5 nm) was formed at a particular site as shown in Fig. 2b. By the repetition of applying voltage, dot structures (width: 20–40 nm, height: 1.5–2.0 nm) were fabricated at the particular sites as shown in Fig. 2c–e.

Surface Structure Transition at 430°C Under As4 Irradiation

In order to increase crystal quality of nano structure, we tried to fabricate at 430°C under As4 irradiation. After the InAs WL growth at 500°C, a substrate temperature has decreased to 430°C under As4 irradiation. When the substrate temperature was stabilized, the STM units were combined and immediately started in situ STM observation. A tip bias was −0.2 V, a tunneling current was 0.4 nA, and the scan speed was 1,000 nm/s. After eliminating the thermal drift, we applied voltage from −0.4 to +0.4 V. White circles shown in Fig. 3 indicate hole structures (width: 15–18 nm, depth: 0.9–1.1 nm), using it as markers for confirming a same position. By the repetition of applying voltage as shown in Fig. 3b, c, five dot structures (width: 16–20 nm, height: 0.75–1.5 nm) were appeared as indicated the white arrows in Fig. 3d. But, these structures were disappeared at the subsequent image shown in Fig. 3e, these only remained at least 40 s and collapsed less than 1000 s. We consider that the collapse of the dot structures were caused by the high mobility of In atoms at this temperature [11–14].

Surface Structure Transition at 300°C Under As4 and In Irradiations

To prevent the collapse of the dots, we try to fabricate at 300°C. After 1.5 ML of the InAs WL growth at 500°C, we stopped In supply and decreased a substrate temperature to 300°C. We first fabricated a hole structures (width: 25 nm,
Fig. 1 In situ STM images of the surface structure transitions on the InAs WL without As$_4$ irradiation. (a–c) White points indicate the positions of applying voltage. (a’–c’) Show after applying voltage from −6 to +6 V at 50°C, −6 to +6 V at 150°C, and −3 to +3 V at 150°C.

Fig. 2 Continuous STM images of surface structure transitions on the InAs WL by applying voltage from −0.5 to +0.5 V without As$_4$ irradiation (substrate temperature: 240°C). White points in (a–d) indicate positions before applying voltage. Dot structures were appeared at particular site in (b–e).
depth: 3 nm) by applying voltage from −1 to +1 V as shown in Fig. 4a. A tip bias was −0.6 V, a tunnel current was 0.3 nA, and the scan speed was 3,000 nm/s. This hole structure which reached to the GaAs substrate might be considered as the Ga-rich sites compared with other InAs WL region [15]. After fabricating the hole structure, we started supplying In flux again. The amount of InAs supply was estimated at 0.02 ML in every scan. After supplying additional 0.02 ML and 0.04 ML of InAs, respectively.

Fig. 3 In situ STM images of surface structure transitions on the InAs WL by applying voltage at the particular sites (substrate temperature: 430°C). White circles indicate hole structures, using it as markers for confirming the same position. a Before, b and c during applying voltages, d dot structures were appeared as indicated by white arrows, and e these dots were collapsed.

Fig. 4 STMBE images of a dot grown in a hole structure under In and As4 irradiations at 300°C and its line profiles. a Shows the hole structure, b and c were images after supplying additional 0.02 ML and 0.04 ML of InAs, respectively.

dot structure in the hole structure and the S−K dots. Figure 5 shows the schematic illustration of the fabrication process of a nano dot growth. By applying voltage, a hole structure was artificially created as shown in Fig. 5b, which dug the WL. This hole structure might be considered as the Ga-rich sites compared with other InAs WL region. The migration barrier for In atoms decreases going from GaAs to InAs [9]. This site congregated In atoms, which were mainly migrating on the WL (Fig. 5c), and went easily beyond its critical thickness, forming the dot structure (Fig. 5d).

Conclusion
Site-controlled InAs nano dots were successfully fabricated by the STMBE system at the substrate temperatures from
50 to 430°C. After 1.5 ML of the InAs WL growth by ordinal S–K dot fabrication procedures, we applied voltage at particular sites on the InAs WL, creating the site where In atoms, which were migrating on the WL, favored to congregate. At 240°C, site-controlled InAs nano dots were fabricated. At 430°C, InAs nano dots were also fabricated. However, these dots were remained at least 40 s and collapsed less than 1000 s. Then, we fabricated InAs nano dots at 300°C under In and As₄ irradiations. These were not collapsed and considered to high crystalline dots. Our results indicated that there was a possibility to grow site-controlled array of InAs QDs or a single QD with high crystalline quality during an MBE growth.

Acknowledgments This work was supported by KAKENHI (22918017).

Open Access This article is distributed under the terms of the Creative Commons Attribution Noncommercial License which permits any noncommercial use, distribution, and reproduction in any medium, provided the original author(s) and source are credited.

References

1. Y. Okada, R. Oshima, A. Takata, J. Appl. Phys. 106, 024306 (2009)
2. H. Sakaki, Jpn. J. Appl. Phys. 28, L314 (1989)
3. A. Luque, A. Marti, Phys. Rev. Lett. 78, 5014 (1997)
4. S. Ohkouchi, Y. Sugimoto, N. Ozaki, H. Ishikawa, K. Asakawa, Appl. Surf. Sci. 254, 7821 (2008)
5. F. Vazquez, D. Sonoda, Y. Miyamoto, K. Furuya, Tech. Rep. IEICE. ED93-118, CPM93-89, 43–48 (1993-11) [in Japanese]
6. M. Mehta, D. Reuter, A. Melnikov, A.D. Wieck, A. Remhof, Physica E 40, 2034 (2008)
7. Y. Nakamura, O.G. Schmidt, N.Y. Jin-Phillipp, S. Kiravittaya, C. Müller, K. Eberl, H. Gräbeldinger, H. Schweizer, J. Cryst. Growth 242, 339 (2002)
8. S. Tsukamoto, N. Koguchi, J. Cryst. Growth 201, 118 (1999)
9. S. Tsukamoto, T. Honma, G.R. Bell, A. Ishii, Y. Arakawa, Small 2, 386 (2006)
10. T. Honma, S. Tsukamoto, Y. Arakawa, Jpn. J. Appl. Phys. 45, L777 (2006)
11. E. Steimetz, J.-T. Zettler, W. Richter, D.I. Westwood, D.A. Woolf, Z. Sobiesierski, J. Vac. Sci. Tech. B 14, 3058 (1996)
12. D.I. Westwood, Z. Sobiesierski, C.C. Matthai, E. Steimetz, T. Zettler, W. Richter, J. Vac. Sci. Tech. B 16, 2358 (1998)
13. E. Steimetz, T. Wehnert, K. Haberland, J.-T. Zettler, W. Richter, J. Cryst. Growth 195, 530 (1998)
14. E. Steimetz, T. Wehnert, H. Kirmse, F. Poser, J.-T. Zettler, W. Neumann, W. Richter, J. Cryst. Growth 221, 592 (2000)
15. T. Toujyou, S. Tsukamoto, Phys. Stat. Sol. (c), (2010) [in press]