Control of Size and Density of Self-organized Quantum Dots Grown on GaAs(311)B by Atomic Hydrogen-assisted Molecular Beam Epitaxy

K. Akahane, T. Kawamura, S. Lan, Y. Okada and M. Kawabe

Institute of Material Science, University of Tsukuba,
1-1-1 Tennodai, Tsukuba, Ibaraki, 305-8573, Japan
Phone/Fax:+81-298-53-6902/+81-298-55-7440, E-mail:akahane@mat.ims.tsukuba.ac.jp

(Received: 15 January 1998; accepted: 2 March 1998)

Abstract

We have investigated methods to control the size and density of self-organized InGaAs quantum dots (QDs) on GaAs (311)B substrates using atomic hydrogen-assisted molecular beam epitaxy, and the formation mechanisms of highly packed QDs have been discussed. It is shown that the QDs size and density can be controlled simply by optimizing the substrate temperature. The average QDs densities have ranged from $5.6x10^9$ /cm^2 for samples grown at 540℃ to $1.4x10^{11}$ /cm^2 for those at 460℃, while the average dot size changed from 20 nm (at 460℃) to 130 nm (at 540℃).

Further, QDs grown on GaAs (311)B have shown remarkably well ordered self-organization behavior compared to those on GaAs (001) substrates. Under a given set of growth conditions, the surface has become highly packed with QDs, and eventually QDs have covered the entire surface with further growth. And yet, QDs have not coalesced to form larger islands as commonly observed for growth on (001) substrates. It is thought that there may exist repulsive force acting in between neighboring QDs associated with phase separation and/or strain energy, which prevents coalescence of QDs.

Introduction

Currently, there is a growing research on the quantum dots (QDs) for the interests of both low-dimensional physics and novel semiconductor quantum effect devices such as single electron tunneling devices and semiconductor quantum wire and dot lasers [1]. The fabrication of self-assembled QDs using Stranski-Krastanov (S-K) islanding growth mode in heteroepitaxy [2-4] is of particular importance because of simplicity and high reliability.

However, it is difficult to obtain uniformly distributed QDs and also to control their positioning on the surface in S-K growth mode, and much efforts have been devoted to solve these problems. Recently, improvements of optical properties have been reported by decreasing the size dispersion with stacked QDs [5]. The control of positioning of QDs has been achieved by adding lithography process before the growth [6]. Further, a new growth mode has been proposed by Temmyo et al. for growth on GaAs (311)B substrates by metalorganic vapor phase epitaxy (MOVPE) followed by post-growth anneal [7].

Previously, we showed that it is possible to fabricate ordered InGaAs QDs by using atomic hydrogen-assisted molecular beam epitaxy (H-MBE) on GaAs (311)B substrates as compared to GaAs (001) substrates [8-10]. In this paper, we investigated methods to control the size and density of self-organized InGaAs quantum dots (QDs) on GaAs (311)B substrates using H-MBE. The QDs size and density were controlled simply by optimizing the substrate temperature. The formation mechanisms of highly packed QDs were also discussed, which are different from the pictures proposed by Temmyo et al. [7].

Experiments

In this work, all the samples were fabricated by H-MBE. The effects of atomic H irradiation on cleaning of GaAs and InP substrates are now widely established [11,12]. In fabrication of QDs, atomic H irradiation is known to improve the uniformity of QDs distribution and optical properties [8-10]. Firstly, GaAs (311)B substrates were cleaned with atomic H at 500℃ in UHV. The H_2 back pressure during surface cleaning and InGaAs growth were $6x10^{-6}$ Torr, which corresponded to atomic H flux of about $1x10^{16}$ /cm^2 s. After atomic H cleaning, a GaAs buffer layer of 300 nm thickness was grown at 580℃ at a growth rate of 1 μm/h. An InGaAs QDs layer was then grown at different substrate temperatures ranging from 460 to 540℃ with a 20℃ interval, at a growth rate of 0.1 μm/h. Reflection high-energy electron diffraction (RHEED) was used to monitor the surface reconstruction and morphology during growth. The surface morphology of each sample after growth was

further studied by atomic force microscope (AFM) in air. The photoluminescence (PL) measurements were carried out at 4.2K with optical excitation using an Ar ion laser and a wavelength of 514.5 nm.

Results and Discussions

Figure 1 show the AFM images for (a) $In_{0.4}Ga_{0.6}As$ QDs, and (b) InAs QDs, grown on GaAs (311)B substrates at 500℃ respectively. The thickness of $In_{0.4}Ga_{0.6}As$ was 8.8 monolayers (ML). A clear difference is evident between the two figures, in which the structure is well-ordered for $In_{0.4}Ga_{0.6}As$ QDs. Though it is possible to control the QDs size by changing In composition, the perfection of ordering as seen in Fig. 1 (a) is lost for the case of InAs as shown in (b).

Figures 2 shows the AFM images of $In_{0.4}Ga_{0.6}As$ QDs grown on GaAs (311)B substrate at; (a) 460, (b) 480, (c) 500, (d) 520, and (e) 540℃, respectively. The AFM scan area is 1x1 μm^2 in each image. It can be observed that QDs size and density can be controlled without distracting their ordered structures. Figure 3 plots the results of average QDs size

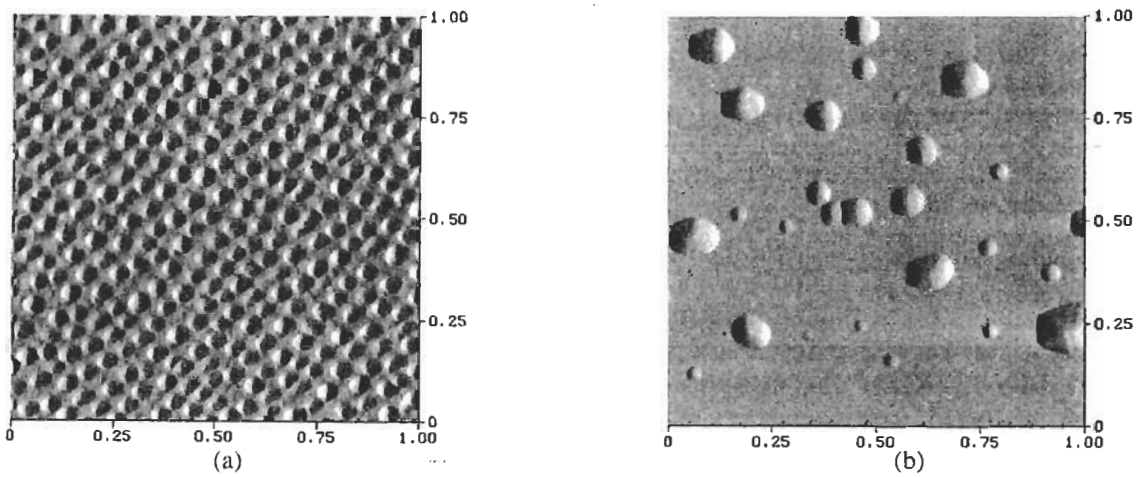

Fig.1 AFM image of grown on GaAs(311)B substrate at 500℃ (a) In0.4Ga0.6As QDs (b) InAs QDs.

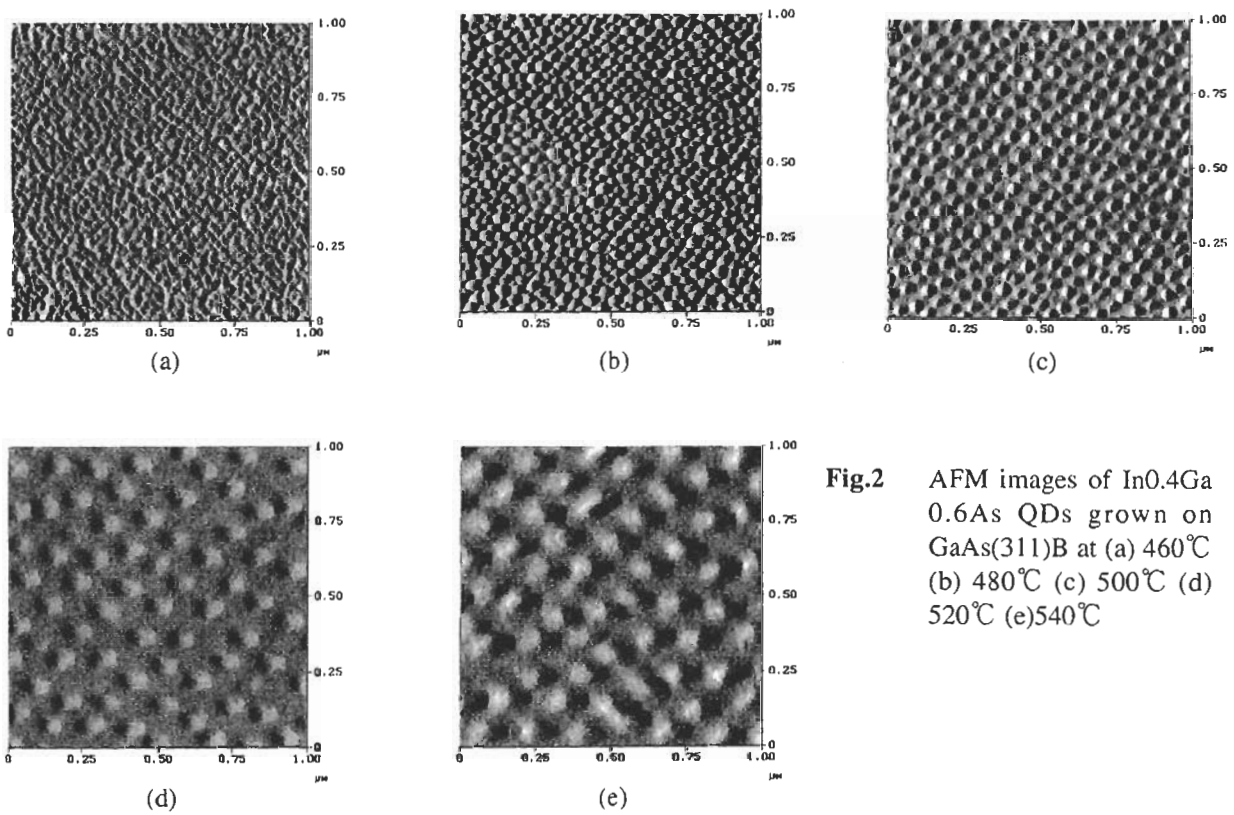

Fig.2 AFM images of In0.4Ga 0.6As QDs grown on GaAs(311)B at (a) 460℃ (b) 480℃ (c) 500℃ (d) 520℃ (e)540℃

(diameter) and density; (a)20 ~ 30 nm, 1.4x 10^{11}/cm^2, (b)30 ~ 40 nm, 8.0x10^{10}/cm^2, (c) 50 ~ 60 nm, 3.3x10^{10}/cm^2, (d) 70 ~ 80 nm, 6.9x10^9 /cm^2, and (e) 110 ~ 130 nm, 5.6x10^9 /cm^2, respectively. In going from (a) to (c), QDs became increasingly highly-packed, and particularly in (a) and (b) the surface coverages of QDs were almost 100%. In addition, QDs did not appear to coalesce to form larger islands as observed in InAs QDs as shown in Fig. 1 (b). These observations were uniquely observed in our low growth temperature MBE, and significantly different from QDs grown by MOVPE on GaAs (311)B [7].

The QDs in Fig. 1 (b) were grown under the same conditions as those of Fig. 1 (a) except the In composition. The $In_{0.4}Ga_{0.6}As$ QDs were packed closely without coalescence, while InAs QDs were more spaciously distributed and larger islands were also seen as a consequence of coalescence of smaller QDs with possible introduction of dislocations [13]. These structures could be explained in terms of possible existence of repulsive force acting in between neighboring QDs associated with phase separation and/or strain energy, which would act to prevent coalescence of QDs and result in featured ordering. This is because InGaAs ternary is found to be energetically unstable around In composition of 0.4 due to a miscibility gap [14,15], and hence phase separation is expected to occur during MBE growth at low temperatures. If phase separation occurs in the QDs, then an In-rich phase would be located above a Ga-rich phase, as schematically shown in Fig. 4, because it would relax the lattice strain energy in the most efficient way.

Further, Fig. 5 shows the PL spectra measured from the samples grown at (a) 460, (b) 480, (c) 500, and (d) 520℃, respectively. The peaks at around 1.5 eV were from GaAs buffer layer, and peaks at 1.49 eV were attributed to carbon-acceptor transitions. The PL peak energy and full-width at half maximum (FWHM) were; (a) 1.27 eV, 21 meV, (b) 1.31 eV, 33 meV, (c) 1.32 eV, 40 meV, and (d) 1.28 eV, 45 meV, respectively. PL peak intensity seen at 1.28 eV in sample (d) was significantly low compared to (a) through (c). The reason is considered to be that QDs are less in density and larger in size for the high temperature sample (d) compared to those of (a) through (c), and thus it is thought that either dislocations are introduced in (d) during cooling process from growth temperature and/or QDs are degraded during the growth of cap layer at a high-temperature of 520℃. We believe further studies are necessary to optimize the growth conditions of cap layers. There is a larger red shift of the PL peak in (a) compared to (b), (c), though the QDs are smaller in size in (a) and hence the expected quantum confinement would be larger for (a) than (b), (c). Lateral optical coupling may account for these results in a similar fashion to vertical coupling reported for stacked QDs. Finally, PL peak intensity from sample (a) was as large as that for (b), (c), which indicated the lower temperature sample (a) is of almost the same optical quality as (b), (c).

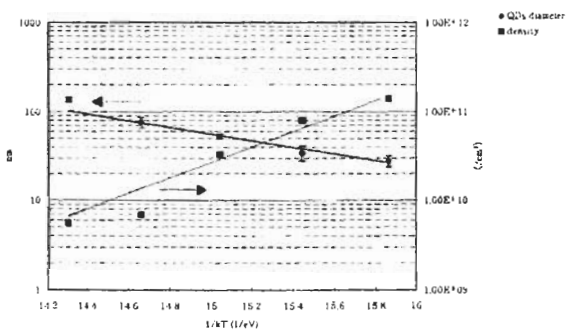

Fig.3 Variation of dot diameter and density of $In_{0.4}Ga_{0.6}As$ quantum dots on GaAs(311)B with growth temperature.

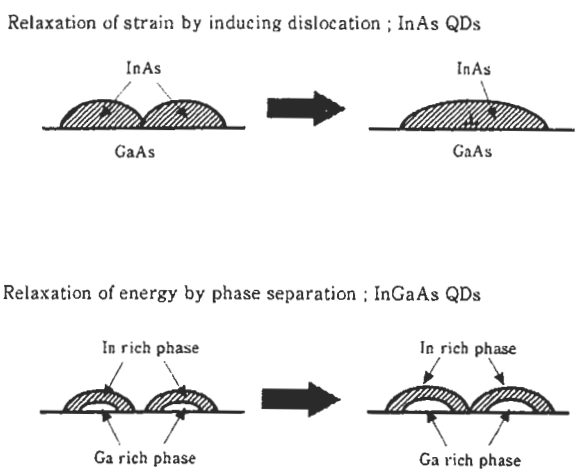

Fig.4 The schematics of the model for QDs configuration change (a) InAs QDs (b) InGaAs QDs.

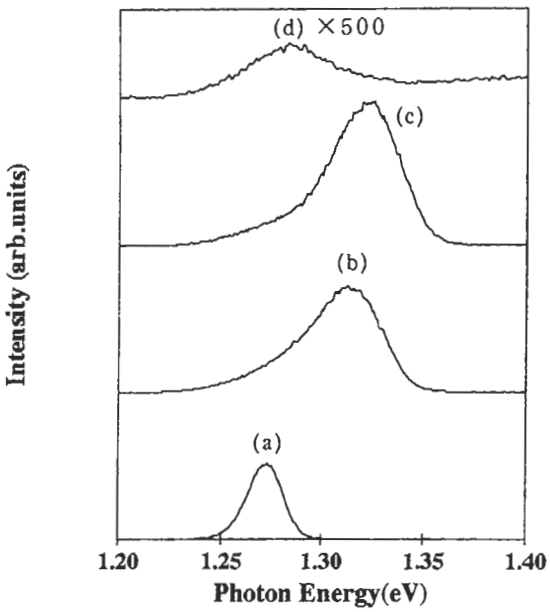

Fig.5 PL spectra of $In_{0.4}Ga_{0.6}As$ QDs samples grown on GaAs(311)B at (a) 460℃, (b) 480℃, (c) 500℃, (d) 520℃respectively.

Conclusion

We showed a method to control the QDs size and density grown on GaAs (311)B substrates by simply optimizing the substrate temperature in H-MBE technique, without destruction of QDs ordering. The $In_{0.4}Ga_{0.6}As$ QDs were closely packed without coalescence, while InAs QDs were more spaciously distributed and larger islands were also seen as a consequence of coalescence of smaller QDs. These structures were explained with a model based on repulsive force acting in between neighboring QDs associated with phase separation and/or strain energy, which would act to prevent coalescence of QDs and result in featured ordering. Furthermore, the PL results indicated not only the optical qualities of these QDs, but some important quantum characteristics were observed which will be discussed elsewhere.

Acknowledgments

We acknowledge the Cryogenic Center of the Unimersity of Tsukuba for use of the equipment for photoluminescence measurements.

Reference

1. Y.Arakawa and H.Sakaki, Appl. Phys. Lett. 1982, **40**, 939
2. L.Goldstein, F.Glas, J.Y.Marzin, M.N.Charasse and G. Le Roux, Appl. Phys. Lett. 1985, **47**, 1099
3. D.Leonard, M.Krishnamurthy, C. M. Reaves, S. P. Denbaars and P.M.Petroff, Appl. Phys. Lett. 1993, **63**, 3203
4. R.Nözel, J.Temmyo and T.Tamamura, Nature 1994,**396**,131
5. Y.Sugiyama, Y.Nakata, T.Futatsugi, M.Sugawara, Y.Awano and N.Yokoyama, Jpn. J. Appl. Phys. 1997, **36**, L158
6. E. Kuramochi, J. Temmyo, and T. Tamamura, Appl. Phys. Lett. 1997, **71**, 1655
7. J.Temmyo, R.Nözel and T.Tamamura, Appl. Phys. Lett. 1997, 71, 1086
8. Y.J.Chun, S.Nakajima, Y.Okada and M.Kawabe, Physica B 1996, **227**, 299
9. Y.J.Chun, S.Nakajima and M.Kawabe, Jpn. J. Appl. Phys. 1996, **35**, L1075
10. M.Kawabe, Y.J.Chun, S.Nakajima and K.Akahane, Jpn. J. Appl. Phys. 1997, **36**, 4078
11. T.Sugaya and M.Kawabe, Jpn. J. Appl. Phys. 1991, **30**, L402
12. Y.J.Chun, T.Sugaya, Y.Okada and M.Kawabe, Jpn. J. Appl. Phys. 1993, **32**, L287
13. C.W.Snyder, B.G.Orr, D.Kessler and L.M.Sander, Phys. Rev. Lett. 1991, **66**, 3032
14. K.Onabe, Jpn. J. Appl. Phys. 1982, **21**, L323
15. G.B.Stringfellow, J. Appl. Phys. 1983, **54**, 404