Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Journal of Crystal Growth 278 (2005) 342–345
JOURNAL OF CRYSTAL GROWTH
www.elsevier.com/locate/jcrysgro

Regrowth dynamics of InAs quantum dots on the GaAs circular mesa

Zhigang Xie[a], Fang Wei[b], Hui Cao[b], Glenn S. Solomon[a,*]

[a]Solid State Photonics Laboratory, Stanford University, Stanford, CA 94305, USA
[b]Department of Physics & Astronomy, Northwestern University, Evanston, IL 60208, USA

Abstract

We report MBE regrowth of InAs quantum dots on GaAs circular mesas, prepared by optical lithography. Because of better strain relaxation, the possibility of quantum dots growth near the lithographic edge is high. Under controlled growth conditions, quantum dots appear only close to the edge. Under these conditions, we discuss the possible influence of crystal orientation to the quantum dots formation, as well as geometrical factors, such as the lateral size of the mesa, and the depth and steepness of the lithographic step. With the full control of the quantum dots formation, we measured the photoluminescence spectrum of the buried dots, as well as the real space image from a CCD camera. The results indicate that quantum dots only form at the edge. Besides the physical location, all the other parameters are quite similar to self-assembled quantum dots formed on planar surfaces.

PACS: 68.65.Hb; 68.65.−k; 81.07.Ta; 81.07.−b

Keywords: A1. Diffusion length; A2. Regrowth; A3. Quantum dots; B1. InAs

Because of the atom-like nature, self-assembled quantum dots (QD) are of great interest as a single photon emitter [1,2]. From early 1990s, this strain-driven phenomenon has been realized and widely investigated on many lattice-mismatched systems, such as Ge/Si [3], InAs/GaAs [4] and InGaAs/GaAs [5]. Despite the nature of random distribution, some techniques are used to "guide" the QD formation. One of them is a QD superlattice [6], in which the underlying layer of QDs serves as the stressor of the upper-level QDs. Another approach is to use lithographic features to seed the QD formation or confine the lateral size of QDs [7]. In this case, usually ultra-high-resolution lithography is required to define the sub-100 nm QDs. In this paper, we will discuss another way to control QDs positioning without the aid of ultra-high-resolution lithography, that is, self-assembled QDs formation on an optical lithographic edge. Kamins

*Corresponding author.
E-mail address: solomon@stanford.edu (G.S. Solomon).

0022-0248/$ - see front matter © 2005 Published by Elsevier B.V.
doi:10.1016/j.jcrysgro.2005.01.062

et al. [8] presented a method of Ge QDs formation on Si pixel lines. It may seem trivial, but this approach can be further developed to incorporate QDs into optical devices [9]. In this paper, we present the research of InAs QDs regrowth on GaAs (0 0 1) substrate. This self-assembled prototype system is mostly widely investigated and unlike the Ge/Si system, we can extract the growth dynamics information using optical methods, besides normal material characterizations.

The GaAs (0 0 1) substrate was patterned with normal optical lithography. Circular features from 2 to 20 μm were transferred to the sample using a wetting etching recipe of diluted H_3PO_4:$H_2O_2 = 1:1$ and photoresist was stripped. After standard solvent cleaning, a wetting etching of diluted NH_4OH:$H_2O_2 = 1:1$ was used to remove a thin layer of GaAs (~40 nm). The sample was then transferred to the MBE chamber for growth. After 1-h high-temperature annealing, a buffer layer of 35 nm GaAs, 1.9 ML InAs were sequentially grown on the substrate at a temperature of 500 °C. At this growth temperature, we typically get an area density of 10 QDs/μm^2, and we can see from Fig. 1a that typically QDs only show up close to the lithographic edge. Here, material migration plays an important role in the QD preferential formation. At our growth conditions, with a substrate temperature of 500 °C and an As/Ga flux ratio of 30, a diffusion length on the order of 1 μm can be estimated. Hence, for the sizes of the circular mesas used, we see no substantial QD distribution in the center area of the mesa. With an increasing of InAs coverage, we noticed that QDs first fill up the edge before they appears elsewhere. For comparison, an AFM image of QDs distribution at a growth temperature of 480 °C is presented in Fig. 1b. We can see that QDs appear almost everywhere, but the density decreases from the edge to the center. So with a diffusion length of tens of nanometers, it is less obvious that the QDs are aligned with the lithographic edge.

Besides the diffusion length, the sidewall of the mesa plays an important role in the QD formation. The InAs material will not only migrate from the top of the mesa but also from the sidewall. To prove this, we apply the QD linear density as a "gauge" to measure the InAs migration. We prepared two samples: sample A with an etching depth of 500 nm and sample B with an etching depth of 1 μm. If InAs does migrate from the sidewall, we expect to see a linear density increase with a shrinking of the mesa sizes. We notice that it is true on both samples A and B (Fig. 2). When both the sidewall and top surfaces are present and

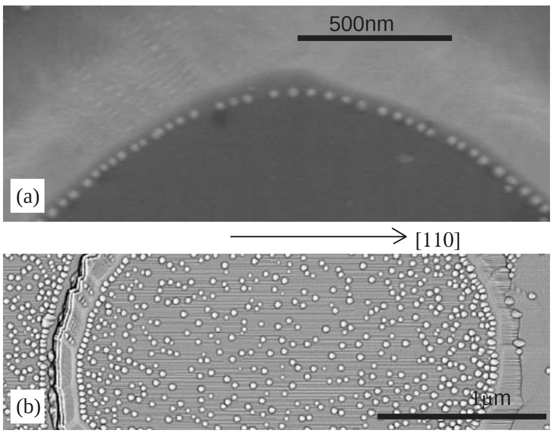


Fig. 1. MBE regrowth on circular mesa structures. (a) The growth temperature is 500 °C. QDs only appear at the lithographic edge. (b) The growth temperature is 480 °C, just 20° lower, and QDs show up on the whole top surface with the area density lower at the mesa center.

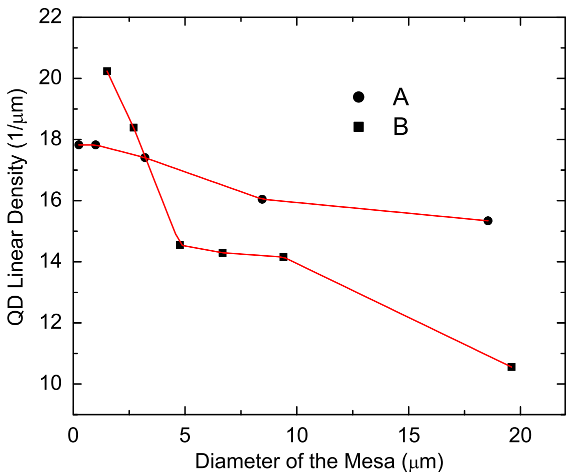


Fig. 2. Average linear density of QDs at the mesa edge. On sample "A", the etching depth is about 500 nm, while on sample "B", the etching depth is 1 μm. On both samples, the linear density slightly increases with the decrease of the mesa diameter.

the In adatoms migrate faster on the side wall ((1 0 1) surface for example) than the top surface ((0 0 1) surface), we will see InAs segregation on the top surface close to the edge. All the above analyses are based on an isotropic distribution. However, the migration could also depend on the crystal orientation. In Fig. 3 we show the QDs distribution for four different crystal orientations ([1 0 0], [0 1 0], [$\bar{1}$ 0 0] and [0 $\bar{1}$ 0]). We can see that there is a special crystal orientation ([1 0 0]), where QDs will also appear in the vicinity of the edge.

To make clear the role of the sidewall for the diffusion of In adatoms, we design another series of samples beginning with 100 nm of GaAs on top of a 600 nm $Ga_{0.3}Al_{0.7}As$ layer. In contrast with the samples described above, we apply a selective etch to make an undercut after the nonselective etch of H_3PO_4:H_2O_2.. In this case, we make disk-like structures instead of the circular mesa. The purpose of the undercut is to determine the sidewall contribution to InAs migration. With the fixed GaAs top layer (a thickness of 100 nm) and same kind of sidewall tilt, we see a big difference in the QD density between the small and big disk samples. These results are discussed in Ref. [9]. On small disk sample (~3 μm), we obtain a linear density as low as 0.5 QDs/μm. This further supports our conclusion that the In sidewall contribution is at least as important as the material migration from the top surface in mesa structures (without undercuts).

For the above comparison between disk and mesa samples, the tilt of the sidewall is the same (very close to 45°). In our definition 90° means a completely straight sidewall. However, it is possible to adjust the sidewall tilt using chemical etching and MBE growth. Fig. 4 shows how the QD density depends on the sidewall tilt. With the same top layer thickness of 100 nm, the lateral widths of the sidewall are 80 nm in (a), 150 nm in (b) and 600 nm in (c). The disk diameters are 20 μm for all three cases. We can see that the QD

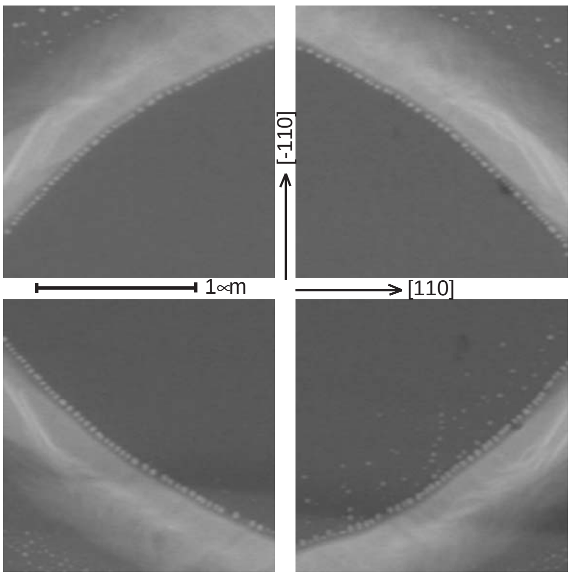


Fig. 3. The QD linear density dependence on the crystal orientation. The QDs distribution is almost uniform along the mesa edge. However, owing to non-inversion symmetry, QD alignment with the edge is relatively poor between [1 1 0] and [1 $\bar{1}$ 0] direction.

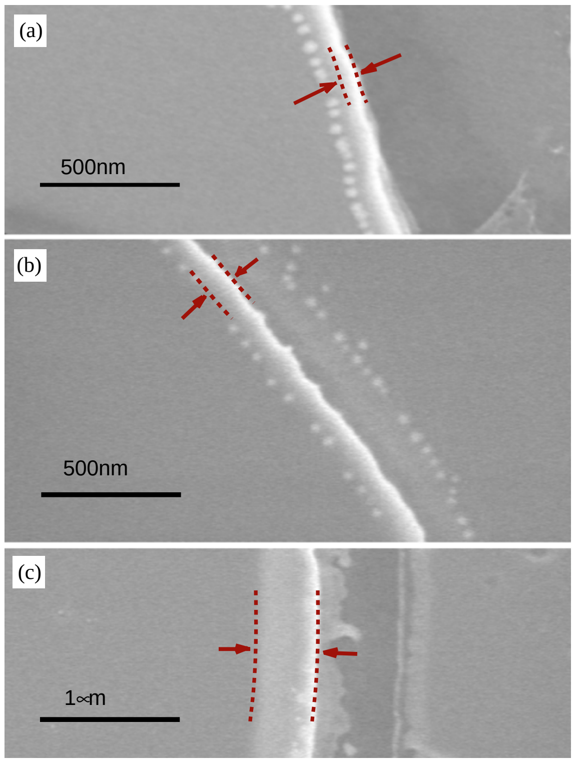
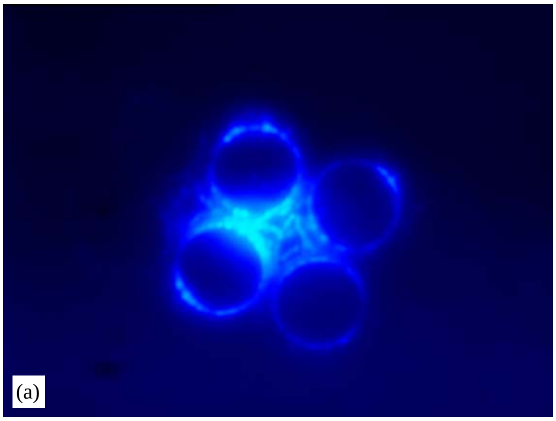


Fig. 4. The QD linear density dependence on the tilt of the sidewall. The sidewall is about 80 nm wide in (a), 150 nm wide in (b) and 600 nm wide in (c). For all three samples with undercut, the GaAs top layer is about 100 nm thick.



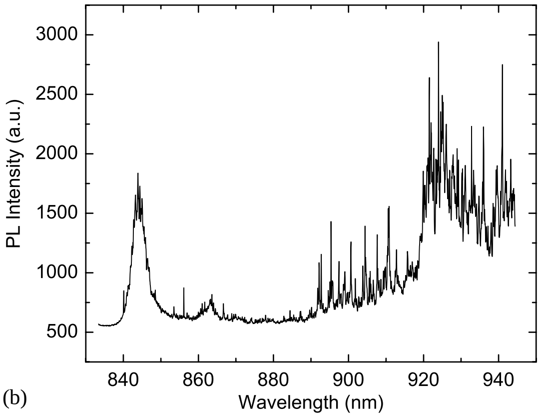


Fig. 5. (a) The CCD image of PL from several 15 μm disk samples very close to each other. (b) A typical PL from the edge of a 25 μm disk sample, the collection area is limited to 5 μm. According to AFM or SEM observations, there should be ~50 QDs in the spectrum range of 890–940 nm.

density is highest in (a), a bit lower in (b) and almost no dots in (c) while the growth conditions are the same. In case (c), instead of providing In adatom flux, there will be a net in-flow of In adatoms because of better strain relaxation on (0 0 1) vicinal surfaces.

With the complete control of QD growth, we capped the QDs with an additional 100 nm of GaAs layer and make optical measurement. The sample was installed in an angle-pumped and He-flow cryostat. The projection of a pinhole limits the photoluminescence (PL) to that from a 5 μm wide region. Fig. 5a shows a CCD image of several spatially very close disk sample. On top of the disk, only PL from the edge is observed. However in between the disks, occasionally we also see the QDs PL emission from the bottom, but far away from the edge. Fig. 5(b) shows a typical PL spectrum from a 25 μm disk sample. The wetting layer signal is at 850 nm, while QDs signals are between 890 and 940 nm and the sharp peaks corresponding to signals from individual QDs.

In summary, we have discussed the growth conditions necessary to place QDs at the lithographic edge of patterned structures. The regrowth mechanism is explored in experimental detail. Besides the influence of the mesa size and crystal orientation on the QD density, we discussed the role of the sidewall including the length and tilt. With these geometric factors as well as growth parameters, it is possible to control the QDs density in a useful optical device such as a microdisk cavity.

We would like to thank Prof. Y. Yamamoto and J.S. Harris for using the lab equipment and Dr. C. Santori for helpful discussions about this project.

References

[1] P. Michler, A. Kiraz, C. Becher, W.V. Schoenfeld, P.M. Petroff, L.D. Zhang, E. Hu, A. Imamoglu, Science 290 (2000) 2282.
[2] C. Santori, D. Fattal, J. Vuckovic, G.S. Solomon, Y. Yamamoto, Nature (London) 419 (2002) 594.
[3] J. Tersoff, C. Teichert, M.G. Lagally, Phys. Rev. Lett. 76 (1996) 1675.
[4] Y. Ebiko, S. Muto, D. Suzuki, S. Itoh, K. Shiramine, T. Haga, Y. Nakata, N. Yokoyama, Phys. Rev. Lett. 80 (1998) 2650.
[5] J. Oshinowo, M. Nishioka, S. Ishida, Y. Arakawa, Appl. Phys. Lett. 65 (1994) 1421.
[6] G.S. Solomon, J.A. Trezza, A.F. Marshal, Phys. Rev. Lett. 76 (1996) 952.
[7] J. Robinson, O. Dubon, J. Alexliddle, A. Minor, 27th International Conference on the Physics of Semiconductors, Flagstaff, Arizona, USA, 2004.
[8] T.I. Kamins, R. Stanley Williams, Appl. Phys. Lett. 71 (1997) 1201.
[9] Z.G. Xie, G.S. Solomon, unpublished.