Formation of highly anisotropic GaAs quantum dots on GaAs(001) substrates

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Abstract. We report on the formation of highly anisotropic GaAs quantum dots on GaAs (001) substrates using droplet epitaxy. Uncapped annealing of the dots brings significant elongation along the [1 10] azimuth, resulting in highly anisotropic GaAs quantum dots. Effects of structural anisotropy of the quantum dots are studied in terms of polarized emission.

1. Introduction

Nanostructure self-assembly has been extensively studied from fundamental as well as technical points of view [1]. In order to derive the desired physical properties it is important to control the morphology of the nanostructure. For example, highly symmetric quantum dots (QDs) are believed to play a crucial role in suppressing the exciton fine-structure splitting that is a major obstacle to producing entangled photon pairs from the QDs [2, 3]. In contrast elongated QDs or quantum dash is another interesting nanostructure which, due to the preferential elongation, allows the realization of polarization-sensitive functionalities such as vertical-cavity surface-emitting lasers with distinct polarization states [4, 5].

So far, growth of quantum dashes has been demonstrated in lattice-mismatched systems such as In(Ga)As/InP [6-8] and InGaAs/GaAs [9], in which the dots spontaneously elongate in order to minimize the total surface/interface energy and elastic strain energy of the epitaxial islands [10]. In such strained systems, however, pure geometric confinement is greatly modified by strain-induced potential. This complicates a straightforward clarification of the effects of structural anisotropy on the physical properties of the nanostructures. As an alternative self-assembling method, droplet epitaxy (DE) has been developed in lattice-matched systems to produce various nanostructures [11-15]. In self-assembled GaAs/AlGaAs nanostructure grown by DE, size and shape of the nanocrystals are dependent on the As pressure as well as on the temperature in crystallizing Ga droplets. For example, dots are formed under intense As flux while ring-like shapes are obtained at relatively low As supply [14]. In both cases, however, the shape of the nanocrystals grown at low temperature has so far been isotropic in plane.

Here, we report on the formation of highly anisotropic GaAs QDs grown on GaAs (001) substrates using DE. Uncapped annealing induces the QDs elongation along the [110] azimuth, which is ruled by the anisotropy in surface step energy.
2. Experimental

Samples were grown on semi-insulating GaAs (001) substrates by molecular-beam epitaxy. After the growth of a 100-nm Al\textsubscript{0.35}Ga\textsubscript{0.65}As buffer layer at 580°C, 1.5 monolayers (MLs) of Ga were supplied to form Ga droplets with a flux of 0.1 ML/s at 300°C. Next, the droplets were crystallized into GaAs under As\textsubscript{4} flux irradiation of 2.5×10\textsuperscript{-5} Torr at 200°C. Then, the samples were annealed for 10 min under As\textsubscript{4} supply (1.0×10\textsuperscript{-5} Torr) without capping, at temperatures ranging from 350°C to 500°C. During the annealing stage, the surface states were monitored by reflection high-energy electron diffraction (RHEED). Atomic force microscopy (AFM) was used to characterize the surface morphology of the open dots. Optical measurement was performed on the samples capped with a 50-nm Al\textsubscript{0.35}Ga\textsubscript{0.65}As layer followed by a 10-nm GaAs protective coat. The Al\textsubscript{0.35}Ga\textsubscript{0.65}As capping layer was grown in two stages. First, a 40-nm-thick Al\textsubscript{0.35}Ga\textsubscript{0.65}As layer was grown at the same annealing temperature of the GaAs QDs, and then the rest of the Al\textsubscript{0.35}Ga\textsubscript{0.65}As layer was deposited at 580°C. A rapid thermal annealing process (800°C, 4 min.) was maintained for all capped samples to improve the optical quality. Photoluminescence (PL) spectra of the samples were taken at 6 K, using the 532-nm line of a frequency-doubled Nd:YAG laser. The PL signals were dispersed by a monochromator and detected by a cooled Si charge-coupled device array.

3. Results and Discussion

Figure 1 shows AFM images of the uncovered sample surfaces annealed at various temperatures: (a) 350, (b) 400, (c) 450, (d) 475, and (e) 500°C. The base length along [110] and [1\text{-}10] of the dots obtained from the AFM scans are plotted in Fig. 2.

![AFM images](image1)

**Figure 1.** 1×1 μm\textsuperscript{2} AFM images of GaAs QDs annealed at different temperatures: (a) 350, (b) 400, (c) 450, (d) 475, and (e) 500°C. Colour scale (z height) differs for each scan: 20 nm for (a) and (b), 8 nm for (c) and (d), and 3 nm for (e).

![Graph](image2)

**Figure 2.** QD base length along [\text{\bar{1}}10] direction plotted against the base along [110] direction for different annealing temperatures. The line denotes an aspect ratio of 1.0. Note that data at 400°C almost overlap those at 350°C.
As seen in figure 1, the dots become laterally anisotropic with increasing the annealing temperature, while the dot height decreases monotonically. At temperatures lower than 400°C, the dot shape is almost isotropic in plane, reflecting the isotropy of seeding Ga droplets. Then, the base length parallel to [110] starts to increase at temperatures higher than 450°C, whereas the other base length along [110] remains nearly constant over the entire temperature range, which is clearly seen in figure 2. Finally, at 500°C annealing, the mean aspect ratio of the long axis to the short axis reaches ~ 3.4.

Highly anisotropic QDs are formed as a result of the rearrangement of constituent atoms on an anisotropic GaAs(001)-(2×4) surface at high-temperature annealing [16,17]. Figure 3 compares RHEED patterns of the surface after annealing at (a) 400 and (b) 450°C. At 400°C, the surface shows a clear c(4×4) reconstruction with a spotty feature due to the QDs. Then the reconstruction changes to a complex structure at 450°C, which is observed at the transition regime from c(4×4) to (2×4). This is consistent with the temperature at which the elongation of the QDs starts to occur, as confirmed by AFM observations.

Figure 3. RHEED patterns of the annealed surface (a) 400 and (b) 450°C, taken along the [110] azimuth.

The shape of the GaAs QDs is governed both by kinetic and thermodynamic aspects. Kinetically, different surface diffusion constants and sticking coefficients will produce the anisotropic islands. Thermodynamically, the islands become elongated along low-energy steps to minimize the total free energy. In the GaAs(001)-(2×4) surface, the energy of the A-step, which is parallel to the As-As dimers, is lower than that of the B-step, which is perpendicular to the dimers [18]. In addition, Ga atoms are incorporated more easily at the B-steps than at the A-steps [19]. Thus, the GaAs islands on the GaAs(001)-(2×4) surface elongate along [110].

As a result of the shape transition, emission from the QDs becomes polarized in plane. Figure 4 depicts PL spectra of a 500°C-annealed sample with the polarization field along [110] and [110]. The PL intensity is larger in [110] than in [110], illustrating the lateral confinement anisotropy. In the inset, the degree of polarization (DOP), defined as \[ \text{DOP} = \frac{I_{[110]} - I_{[\overline{1}10]}}{I_{[110]} + I_{[\overline{1}10]}} \], where \( I_{[110]} \) is the PL intensity parallel to the [110] ([110]) direction, is plotted as a function of the annealing temperature. As can be seen, the DOP increases in general with increasing shape anisotropy due to heavy- and light-hole mixing [20]. Considering the relatively large size of the QDs grown in this study, DOP of 10% in the 500°C-annealed sample is within a reasonable range compared to that of GaAs/AlAs quantum wires on (775)B [21]. By narrowing the width of the QDs, further enhancement of DOP can be expected.
4. Conclusion
We have reported the self-assembly of highly anisotropic GaAs QDs on GaAs (001) substrates using droplet epitaxy. Thermal annealing at higher temperature than 450°C causes significant dot elongation along [110], accompanied by a decrease in dot height. Change in the surface reconstruction from c(4×4) to (2×4) is considered to be responsible for this shape transition. Emission from the QDs is [110]-polarized as a result of structural anisotropy.

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