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Self-organization of quantum-dot pairs by high-temperature droplet epitaxy

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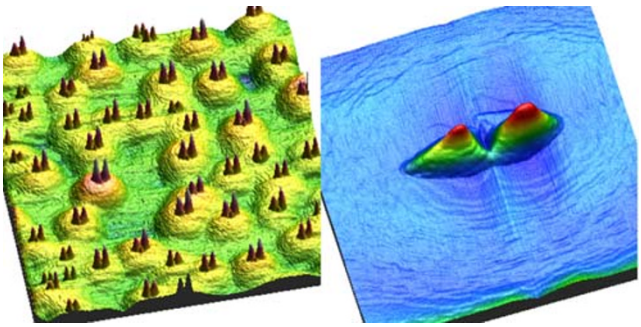
Abstract The spontaneously formation of epitaxial GaAs quantum-dot pairs was demonstrated on an AlGaAs surface using Ga droplets as a Ga nanosource. The dot pair formation was attributed to the anisotropy of surface diffusion during high-temperature droplet epitaxy.



Keywords Quantum dots · Droplet epitaxy

Self-assembly of epitaxial semiconductor nanostructures has been an intensive field of research. In particular, the Stranski-Krastanov (SK) growth mode based on the use of lattice-mismatched materials has played an important role in the formation of nanostructures, the investigation of quantum confinement effects, and has made possible applications of nanostructures [1–3]. While the SK growth mode has been a very powerful and beautiful technique, there has been a significant, but perhaps less well-known, parallel development using lattice matched materials as an alternative approach for the growth of nanostructures called ''droplet epitaxy'' [4–7]. Here liquid metal droplets are first formed as an intermediate growth step before being converted into semiconductor nanostructures. While the two growth approaches are very different, both the SK and the droplet approach are similar in that they both suffer from the stochastic nature of self-assembly. As a result, the control of spatial ordering of semiconductor nanostructures has been extremely challenging while desirable for applications, such as, the fabrication of quantum-dot (QD) molecules for quantum computing [8]. Consequently, there has been much recent effort to control the lateral arrangement of QDs using a range of techniques, such as, lithography [9–12], templating [13, 14], and modified versions of self-organization [15–19]. Recently, the approach of droplet epitaxy has shown promise to the achieve local ordering of quantum nanostructures [6, 7]. In this letter, we report on the use of droplet epitaxy and anisotropic surface diffusion to fabricate QD pairs.

All of the samples used in this study were grown in a molecular beam epitaxy (MBE) system equipped with reflection high-energy electron diffraction (RHEED) and a highly accurate (±2 °C) optical transmission thermometry system for substrate temperature determination and control. Our growth approach was to first grow a 500 nm GaAs buffer layer on a semi-insulating GaAs (100) substrate, followed by a 50 nm thick $Al_{0.3}Ga_{0.7}As$ layer. This was followed by the deposition of Ga and the formation of Ga droplets at the substrate temperature of 550 °C with the arsenic source fully closed. We used a valved arsenic source to provide precise and fast control for over three orders of

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magnitude of the arsenic flux. The total amount of Ga deposited was 10.0 monolayers (MLs), corresponding to the amount of Ga necessary for 10.0 MLs of GaAs growth. A Ga deposition rate of 1.0 ML/s was selected by separately observing RHEED oscillations during the growth of GaAs.

To minimize the system's energy, the deposited liquid Ga forms nano droplets. In particular, by forming nano-droplets the surface to vacuum and interface to the $Al_{0.3}Ga_{0.7}As$ layer, can be minimized, thereby minimizing the system's energy. The atomic force microscopic (AFM) image in Fig. 1a reveals that the Ga droplets have an average lateral size of ~125 nm, a height of ~32 nm, and a number density of ~2.3 per square micrometer. In forming the droplets we have observed that the substrate temperature is a significant parameter for the tuning of droplet density. For example, the density of droplets increases by a factor of six for the same amount of Ga, but deposited at the lower temperature [7] of 380 °C as opposed to 550 °C. We also observed that the size of the droplets could be tuned by controlling the total amount of Ga deposited at a fixed temperature.

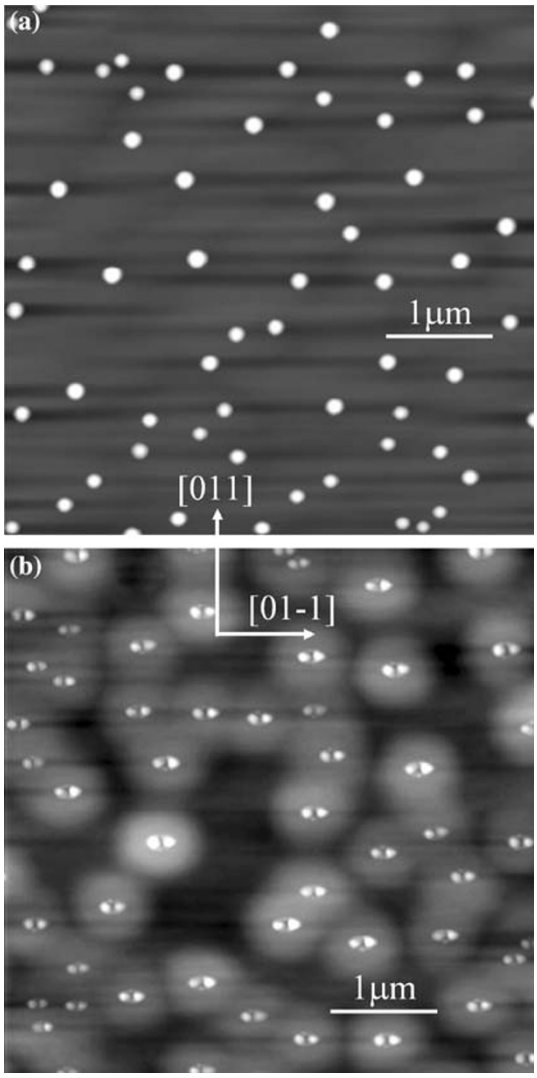

Fig. 1 (**a**) AFM image of Ga droplets. (**b**) AFM image of QD pairs formed after 45 s annealing under arsenic flux

Figure 1b shows an AFM image of the surface nanostructures that are transformed from the Ga droplets after 45 s of annealing while under an arsenic flux of 4.2×10^{-5} Torr. Remarkably, every Ga droplet turns into a QD pair sitting on a 3 nm hill of GaAs. The average separation between two QDs in a pair is about 130 nm, comparable with the original lateral size of Ga droplets. The tips of two QDs are about 4 nm above the GaAs hill.

It is important to note that these results are observed at a substrate temperature of 550 °C which is significantly higher than previously reported droplet-related experiments (growth temperatures of 200 °C or lower) [4–6]. Traditional droplet epitaxy has used low temperatures in an effort to directly crystallize the Ga droplets into GaAs without material redistribution. On the other hand, in our approach we used high-temperature to encourage material redistribution. In this way we can fabricate novel semiconductor nanostructures by taking advantage of the shape instability of Ga droplets during the transformation into GaAs dots. The crystallization of Ga droplets proceeds at a much faster rate at high temperature so that surface processes on the GaAs surface can play a more important role in the shape evolution of the GaAs nanostructure. In particular, the GaAs forms more quickly then it can diffuse away from the droplet. With surface tension removed, the droplet collapses down the center of the droplet pushing material away from the center while restricted by diffusion. Gradually this leaves a smaller and smaller Ga droplet on the GaAs surface acting as a source for forming GaAs diffusing outward, forming ridges and center-holed shaped nanostructures.

In particular, Fig. 2 shows an AFM image of the outcome at high temperature. The AFM image shows that the Ga droplets have fully crystallized after only 1-s of annealing at 550 °C and surface nanostructures shaped like square-holed round coins are immediately observed. Similar shaped nanostructures have been observed for droplet epitaxy performed at 380 °C but with a much longer 45-s annealing and significantly lower arsenic flux [7]. The holed nanostructures are observed to sit on GaAs hills due to the high As flux and corresponding diffusion limited Ga transport. This is confirmed since by lowering the arsenic flux, two-dimensional (2D) growth of GaAs is enhanced and the hills are observed to disappear. As we pointed out

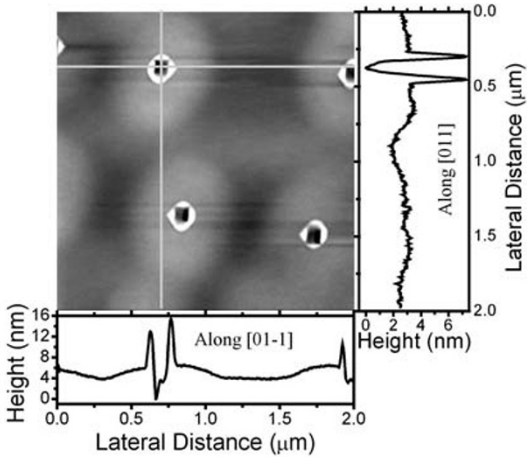

Fig. 2 AFM image of surface nanostructures formed after 1 s annealing and the corresponding height profiles

previously [7], droplet epitaxy can be described as GaAs growth supplied from a Ga nano-source (droplet) under a uniform arsenic flux. From the two line profiles in Fig. 2, the heights of the coin edges are different. In particular, the height is about 9 nm along the [01 – 1] direction and about 4 nm along the [011] direction.

For a typical GaAs (100) surface, it has been well established that surface diffusion and incorporation are anisotropic [20]. With additional annealing, the material from the edges of the square holed nanostructure, tend to fill the center hole but at different rates depending on direction. AFM images in Fig. 3a and b show a step-by-step evolution of the surface of the nanostructures after 5 and 15 s of annealing, respectively. For example, the ratio of the height of the edges along [01 – 1] compared to [011] are seen to increase from Fig. 3a to Fig. 3b forming a bridged pair after 15 s.

Finally, after 45 s of annealing, the bridged nanostructure is encouraged to form a QD pair as shown in Fig. 4a. Additional very small dots indicated by arrows in Fig. 4a are often found to accompany the QD pair. These are the remnants of the edges along [011]. Figure 4b is a 3D presentation of one single QD pair formed after 45 s annealing at 590 °C. At this even higher temperature, the QDs in the dot pairs are bigger and more separated. The clear contour lines around each QD pair are steps on the GaAs hill.

These QD pairs are very stable, as suggested by the observation at different growth temperatures. However, the QD pairs tend to merge together with further annealing.

For example, Fig. 5 shows an AFM image of the QD pair nanostructures after 600 s of annealing.

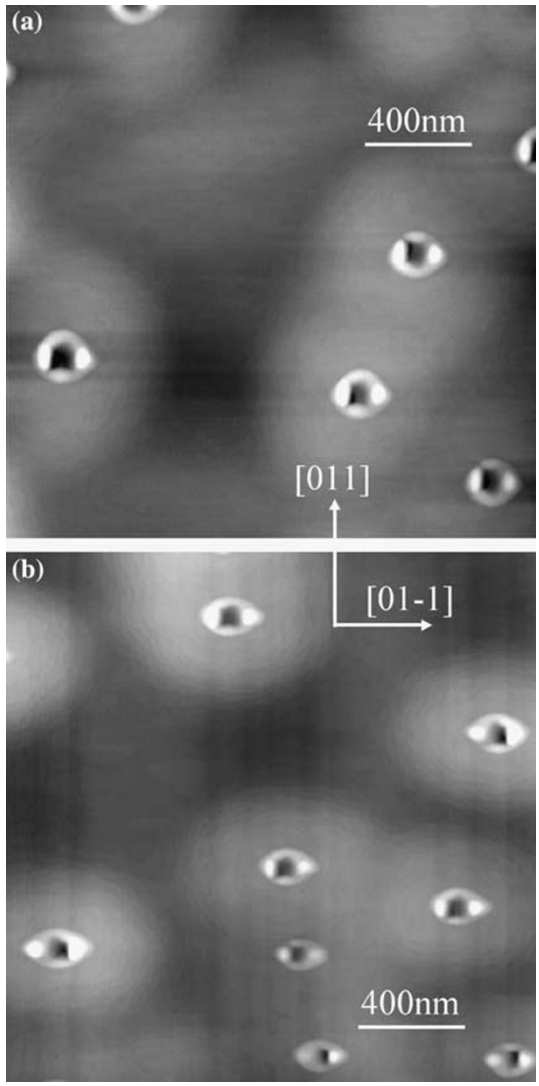

Fig. 3 (**a**) and (**b**) are AFM images of surface nanostructures formed after 5 and 15 s annealing, respectively

Compared to Fig. 4a, the paired QDs have connected together to form a rod sitting on a GaAs hill. The steps on the GaAs hills appear very clear due to the smoothing effects of a long-annealing period. This behavior is not surprising. The 3D nanostructures observed earlier in Figs. 1–4 form from the non-uniform supply of Ga from nano droplets. After the Ga droplets are consumed, additional annealing tends to smear away the nanostructures by surface diffusion and incorporation forming the images in Fig. 5. In this sense, as a result of the anisotropic nature of the crystalline surface, we are able to engineer a variety of different shaped nanostructures ranging from Figs. 1 to 5.

In contrast to the normal droplet epitaxy at low temperatures, high-temperature droplet epitaxy is

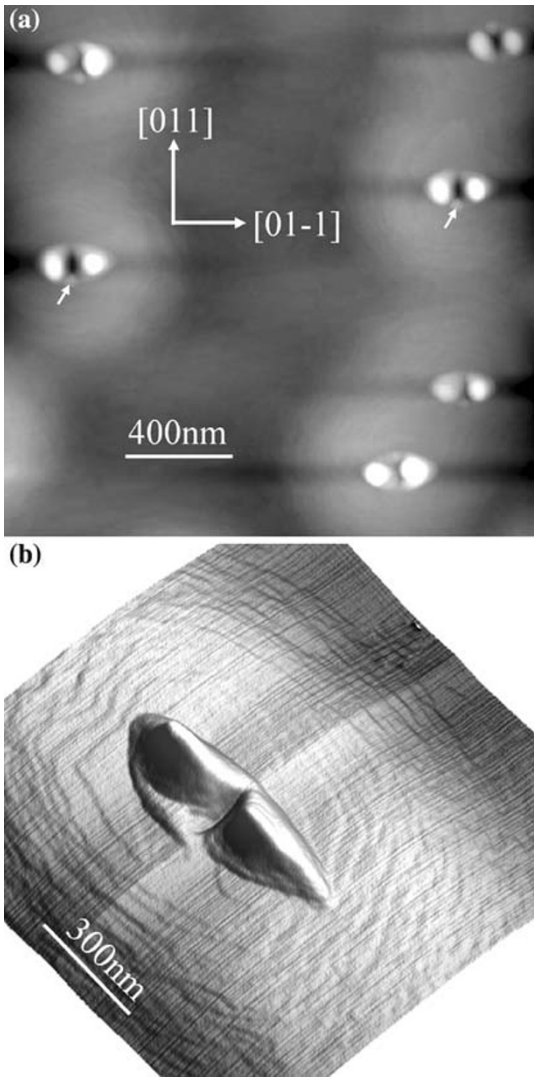

Fig. 4 (**a**) AFM image of QD pairs formed after 45 s annealing. (**b**) A single QD pair formed after 45 s annealing at 590 °C

performed at substrate temperatures very close to normal MBE growth conditions. Therefore, high quality of GaAs/AlGaAs coherent nanostructures with excellent optical properties is demonstrated by near-field scanning optical microscopy. Such studies will be reported in the near future.

In summary, by forming Ga droplets and converting them into GaAs nanostructures at relatively high temperatures, the evolution of QD pairs is observed during subsequent annealing. The dramatic change in shape is caused by the anisotropy nature of surface diffusion and incorporation. High-temperature droplet epitaxy is demonstrated to provide a valuable opportunity to fabricate novel semiconductor nanostructures.

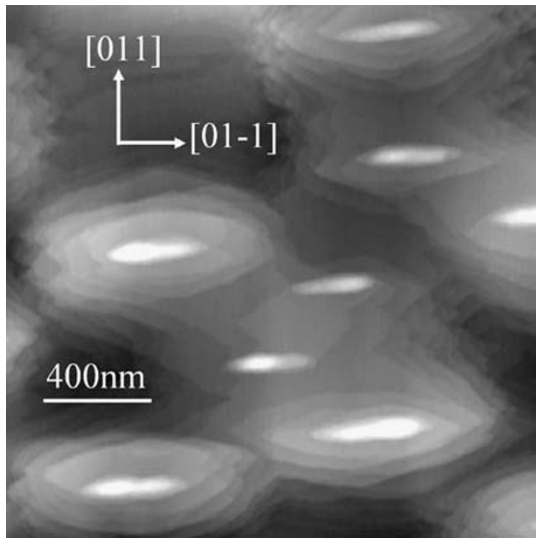

Fig. 5 AFM image of surface nanostructures formed after 10 min annealing

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