Quantum-dot clusters

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Self-Organization of InAs Quantum-Dot Clusters Directed by Droplet Homoepitaxy

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Recently, droplet heteroepitaxy of GaAs/AlGaAs has attracted much attention[1–3] due to its demonstrated capability of producing nanostructures with interesting geometrical shapes such as quantum single and double ringlike structures. In the droplet-heteroepitaxy approach, Ga is deposited on an AlGaAs surface to create liquid Ga droplets, and these droplets are exposed to an As flux that transforms the droplets into GaAs nanocrystals. In principle, this same approach can be modified into a similar process of GaAs homoepitaxy, which also allows one to structure GaAs surfaces at the nanometer scale. In this droplet-homoepitaxy approach, the nanostructures formed by the droplets can then be used to encourage the growth of subsequent heteronanostructures such as self-assembled InAs quantum dots (QDs). In this self-assembly process, the lattice mismatch between InAs and GaAs encourages the spontaneous formation of QDs once InAs deposition reaches a critical thickness.[4–6] These self-assembled InAs QDs have been extensively investigated for the past decade because of their applications in advanced devices such as lasers, infrared detectors, and quantum computers.[6–8] However, the stochastic nature of self-assembly only permits a random distribution of InAs QDs on GaAs surfaces, which presents a major obstacle for applications such as quantum computing. Recently, much effort has been put forth to localize QDs on designed regions of substrates.[9–11] By combining the self-assembly approach with certain lithography techniques,[12–15] InAs QDs can be restricted to growth in specific substrate regions due to GaAs surface height modulations induced by the lithography. In this paper, however, we propose and demonstrate an approach using only self-organizing techniques to localize the QDs, a hybrid process of GaAs droplet epitaxy and strain-driven self-assembly of InAs QDs. By using droplet epitaxy, GaAs(100) surfaces are structured with nanoscale mounds elongated along the $[01\bar{1}]$ direction. The GaAs mounds then induce localized formations of InAs QDs, and these quantum-dot clusters (QDCs) are observed around the GaAs mounds. The density of QDCs and the number of QDs in the clusters can be adjusted by controlling the growth conditions of the Ga droplets. Photoluminescence (PL) measurements reveal atypically good optical properties of these QDCs.

The samples were grown by molecular-beam epitaxy (MBE) on epitaxy-ready GaAs(100) substrates. All growth began with a 300-nm GaAs buffer layer, which was followed by 10 monolayers (ML) of Ga deposition to form the liquid droplets. Subsequently, the Ga droplets were exposed to an As molecular flux and annealed for 2 min at the substrate temperature of 530 °C to fully crystallize the droplets into GaAs surface nanostructures. At the same substrate temperature of 530 °C, InAs was then deposited until the QDs formed after reaching a critical coverage of 1.7 ML. The resulting surface morphologies were imaged by atomic force microscopy (AFM) under ambient conditions. For PL measurements, the resulting InAs QDs were capped with 100 nm of GaAs. Each PL measurement was performed at a temperature of 10 K by using 532-nm laser excitation.

While Ga deposition with a concurrent As molecular flux encourages two-dimensional homoepitaxial growth on GaAs surfaces, Ga deposition without the concurrent As flux results in the formation of liquid droplets, as schematically represented in Figure 1 a. After these droplets subsequently react with the As, they are transformed into GaAs nanostructures, normally taking an anisotropic shape due to the droplets' orientation-dependent surface diffusion on the GaAs crystal surfaces. Figure 1 b shows a schematic repre-

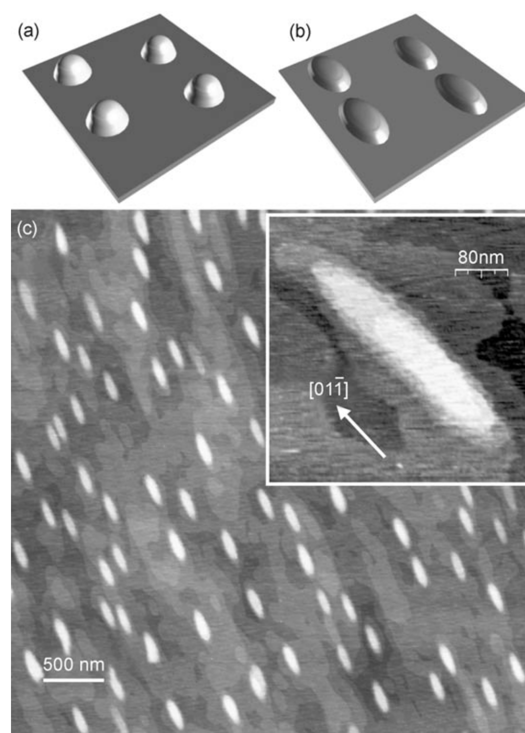

Figure 1. a) Schematic representation of Ga droplets on a GaAs surface. b) Schematic representation of elongated GaAs islands on a GaAs surface. c) 5 × 5 μm² AFM image of the surface topography of the sample when 10-monolayer (ML) Ga droplets were formed at 500 °C, additional annealing took place for 2 min at 530 °C, and 1.4 ML of InAs were deposited. The enlarged AFM image in the inset reveals the atomic layers of the island sides.

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sentation of elongated GaAs mounds, an example of the many possible geometrically shaped GaAs nanostructures that one can create. These nanoscale GaAs mounds form when 10-ML Ga droplets are deposited at 500 °C and annealing takes place for 2 min at 530 °C under an As flux. The surface topography after further deposition of 1.4 ML InAs is shown by the AFM image in Figure 1 c. The density of these surface mounds is about 3.7×10^8 cm^{-2}. The mounds are elongated along the $[01\bar{1}]$ direction and are typically around 440 nm long, 170 nm wide, and 2.4 nm tall, with a length-to-width ratio of 2.6. As shown by the enlarged AFM image in Figure 1 c, the edges of the surface mounds are characterized by ML steps and the density of the steps appears higher on the sides along $[01\bar{1}]$ than the sides along [011].

As shown in Figure 2 a, after the InAs coverage was increased to 1.7 ML, InAs QDs begin to form, but only around the GaAs mounds; this makes them appear as QDCs and distinguishes them from the random distribution of QDs that would normally appear on a flat GaAs(100) surface.[4–6] The average diameters and heights of the QDs are approximately 50 and 12 nm, respectively. The enlarged AFM image in Figure 2 b shows the mounds in greater detail and the ML steps on the surface. The QDs prefer to form on the mound edges along $[01\bar{1}]$ rather than the edges along [011], as schematically represented in Figure 2 c. This can be explained by the mechanism suggested in a recent investigation on shallow-patterned substrates,[15] which indicates that the most favorable regions for formation are in the high-step-density areas. Since the density of ML steps on the mound edges is greater than that of the flat surface, the mound edges become the more favorable sites for the InAs QD formation. In this sample, the statistical distribution of the number of QDs in each QDC is centered at five, as Figure 2 d indicates.

As distinguished from our previous report[15] where outside help from lithography is needed to position the InAs QDs, the hybrid approach from GaAs droplet epitaxy and strain-driven InAs self-assembly is a purely self-organizing technique. While both droplet epitaxy and strain-driven self-assembly are stochastic in nature, the hybrid process produces locally ordered QDCs, groups of QDs surrounding GaAs mounds.

The density and size of the QDCs can be controlled by adjusting the growth conditions of the Ga droplets. Figures 3 a and b show AFM images of the changes in the surface topographies of the GaAs mounds when the Ga droplets are deposited at 550 and 450 °C, respectively. At 550 °C, the density of GaAs mounds approaches 3.1×10^8 cm^{-2} and the mounds appear quite large (780 nm long, 220 nm wide, and 2.3 nm tall). The statistical distribution of the number of QDs in each QDC is centered at ten in this case, as shown in the inset of Figure 3 a. At 450 °C, however, the density of GaAs mounds increases to 5.5×10^8 cm^{-2} and the mounds form with smaller dimensions (358 nm long, 159 nm wide, and 2.2 nm tall) than those formed at 550 °C. Correspondingly, the statistical distribution of the number of QDs in each QDC is centered at two in this case, as shown in the inset of Figure 3 b.

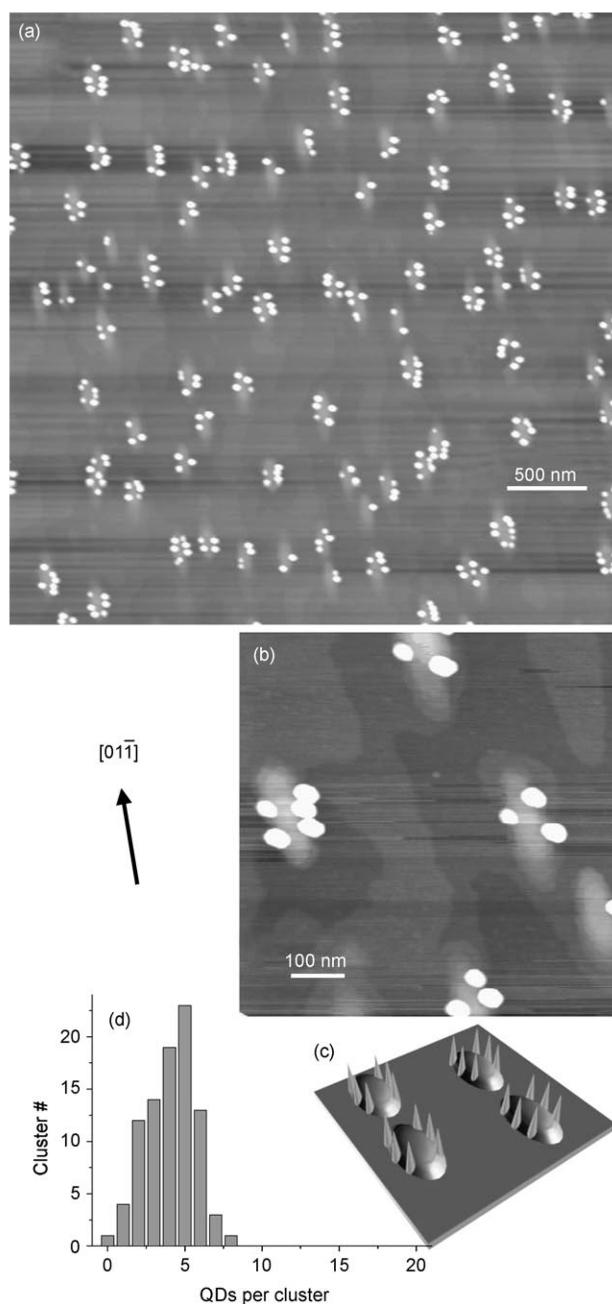

Figure 2. a) 5 × 5 μm^2 AFM image of the surface topography of the GaAs islands formed when Ga droplets were deposited at 500 °C, and InAs QDs were formed after 1.7 ML InAs deposition at 530 °C. b) The 1 × 1 μm^2 AFM image emphasizes the selective formation of the InAs QDs on the sides of the GaAs islands. c) Schematic representation of InAs QDCs induced by GaAs islands. d) Statistical distribution of the number of QDs in the QDCs.

In judging these clustered QDs by typical PL standards, we found them to have good optical properties, similar to most InAs QDs that have been studied, if not better. Figure 4 shows the PL results from capped InAs QDCs produced under the same growth conditions as the sample imaged in Figure 2. With a low excitation power of 0.03 W cm^{-2}, the full-width half maximum (FWHM) of the PL spectrum is less than 33 meV, narrower than the typical

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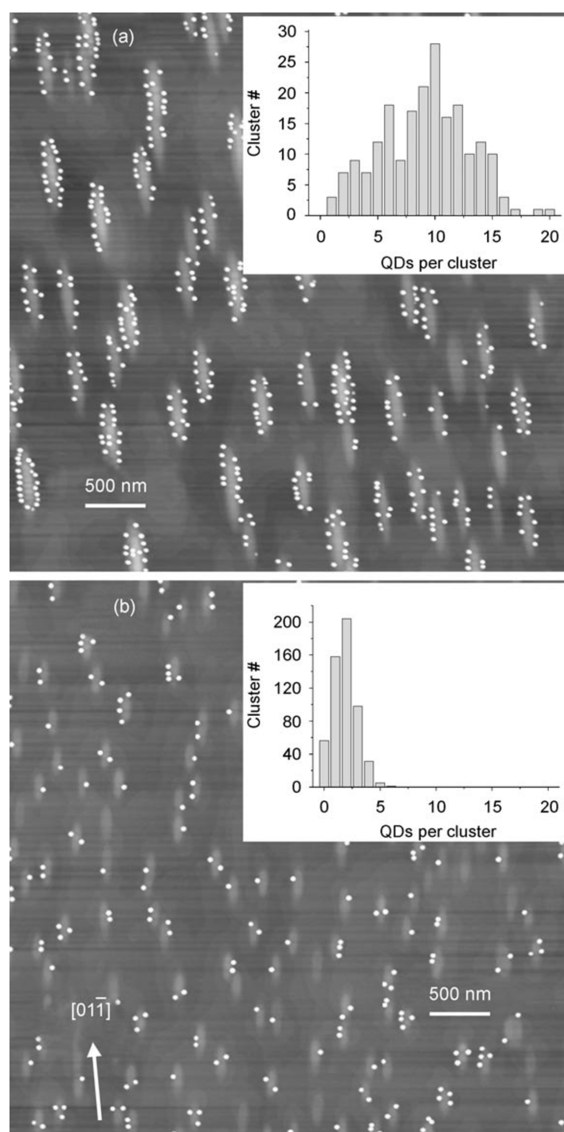

Figure 3. a) and b) $5 \times 5\ \mu m^2$ AFM images of the surface topography with InAs QDs formed after 1.7 ML InAs deposition at 530 °C. (a) shows the GaAs islands formed when Ga droplets were deposited at 550 °C, and (b) shows the GaAs islands formed when Ga droplets were deposited at 450 °C. The corresponding statistical distributions of the number of QDs in the QDCs are shown as insets.

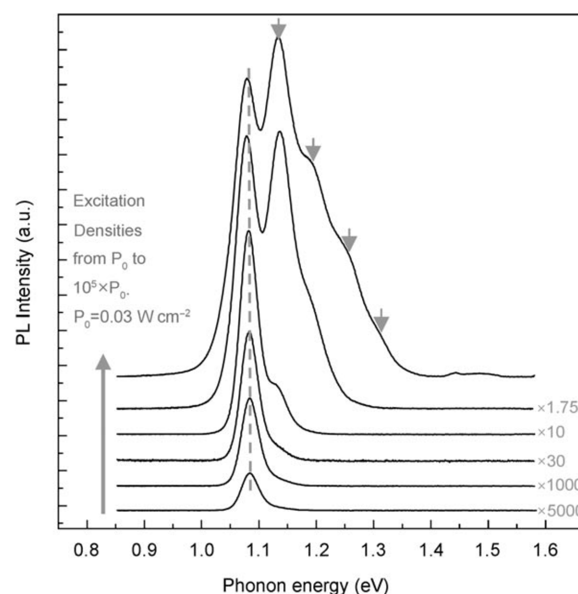

Figure 4. PL spectra of InAs QDCs induced by GaAs islands formed when Ga droplets were deposited at 500 °C. From bottom to top, the six spectra cover the range of laser-power densities from P_0 to $10^5 \times P_0$, where P_0 is 0.03 W cm^{-2}. The dashed vertical line follows the evolution of the PL peak from the ground state, and the short downward arrows point to the PL peaks from excited states.

value reported for InAs quantum dots.[16] With an increase in the laser-power density by five orders of magnitude, the ground-state PL peak, centered at 1.08 eV, shows a negligible shift in energy from the value at the lowest excitation power, but four excited states appear in decreasing order of intensity, as indicated by the short downward arrows in Figure 4. These observed behaviors, namely the narrow PL peak from the ground state, the lack of a shift in energy with increasing excitation power, and the appearance of multiple PL peaks from excited states, indicate that the QDs have desirable optical properties and that the QDs are fairly uniform in size.

In conclusion, by combining techniques from droplet homoepitaxy of GaAs and strain-driven self-assembly of InAs QDs, we have demonstrated a hybrid approach to induce QD formation in localized regions. In the case of droplet homoepitaxy, Ga is first deposited to form Ga droplets on GaAs surfaces, and these droplets then react with an As molecular flux to introduce nanoscale GaAs mounds to the surface. Due to favorable QD formation on high-step-density regions, we are able to limit the subsequent formation of InAs QDs to the sloped portions of the GaAs mounds, as manifested by the clusters of QDs that we observed. By adjusting the growth conditions of the Ga droplets, we have shown how changing the GaAs template affects the growth of the QDCs. Since droplet heteroepitaxy has already been used to grow nanostructures with many interesting shapes, we expect the droplet-homoepitaxy approach to provide more interesting GaAs surface nanostructures (not simply mounds) as templates for subsequent InAs QD growth.[17] Additionally, because of the high optical quality of the InAs QDCs, further modification of the QDC structures may provide an indispensable playing field for basic investigations of carrier interactions among QDs; research in this area of droplet homoepitaxy may yield the desired structures needed for applications in quantum computing.

Keywords:

epitaxy · nanostructures · quantum dots · self-assembly · semiconductors

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