Fabrication of Ultralow Density Quantum Dots by Droplet Etching Epitaxy

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Abstract— Isolated single quantum dots enable the investigation of quantum-optics phenomena for the application of quantum information technologies. In this work, ultralow density InAs quantum dots are grown by combining Droplet Etching Epitaxy and the conventional epitaxy growth mode. The extremely low density of quantum dots (~10^6 cm^{-2}) are realized by creating low density self-assembled nanoholes with the high temperature Droplet Etching Epitaxy technique and then nanohole-filling. The preferred nucleation of quantum dots in nanoholes has been explained by a theoretical model. Atomic force microscopy and photoluminescence technique are used to investigate the morphological and optical properties the quantum dot samples. By varying In coverages, the size of InAs quantum dots can be controlled. Moreover, with a thin GaAs cap layer, the position of quantum dots remains visible on the sample surface. Such a low density and surface signature of quantum dots make this growth method promising for single quantum dot investigation and single dot device fabrication.

Key words— molecular beam epitaxy (MBE), nanostructure, self-assembly

Quantum dots (QDs) are one of the most important nanostructures due to their unique properties and applications in a wide range of novel devices.[1, 2] Since the discovery of QDs, they have been applied to quantum devices[3], solar cells[4], lasers[5], and photodetectors[6]. Fabrication of QDs with user-specified properties is desired for many different applications. For example, a high density of QD desired for solar cells and photodetectors, while a low density of QDs is needed in the fields of quantum computing and quantum cryptography, such as single-photon emitters.[7] In the last ten years, several approaches have been developed to obtain low density of QDs so that each isolated single QD can be addressed optically. After InAs deposition slightly less than the critical coverage value, post-growth annealing is introduced to obtain low density dots around ~10^9 cm^{-2},[8] ~10^8 cm^{-2},[9] and ~10^4-10^8 cm^{-2}.[10] Also, it has been found that low density of QDs can be obtained when InAs is intentionally deposited on the sample surface inhomogeneously.[11] In order to further reduce the quantum dot density, the InAs coverage is intentionally varied along the surface of a highly inhomogeneous sample by stopping the rotation of the wafer during InAs deposition. After additional thermal annealing, QD density as low as 2.5×10^7 cm^{-2} is achieved by using this method.[12] Similarly, InAs is deposited on a GaAs substrate with a temperature gradient. Offcut substrate can also be used to achieve nucleation of low density QDs in the range 10^7–10^8 cm^{-2} without any growth interruption or post-growth annealing.[13] As a result, a QD density gradient is found along the same direction of thermal gradient and QDs with density as low as $\sim 8 \times 10^6$ cm^{-2} are observed in the high temperature region.[14] Recently, very low-density QDs ($\sim 4 \times 10^6$ cm^{-2}) is reported by using a low InAs growth rate and a high growth temperature.[15, 16] Using this simple method, large scale low density QDs can be formed but with poor control over QD position. Pre-patterned substrate has also been used to grow position-controlled low density QDs for potential device integration.[17-19] This method requires pre-growth processing of substrates, which can easily introduce contaminations and defect formation. Additionally, it is difficult to grow low density QDs in large scale limited by the processing techniques.

In this article, we report the fabrication of ultralow density quantum dots formed in self-assembled nanoholes by molecular beam epitaxy. This technique employs the unique Droplet Epitaxy.[20, 21] By using the Droplet Epitaxy, the formation of large scale low density quantum dots can be easily obtained and well controlled. By using high temperature Droplet Etching Epitaxy, the nanohole density and hence quantum dot density is reduced to ~4× 10^6 cm^{-2}.[22] Such a low density of quantum dots provides the opportunities for fabrication of low density QDs and researches on individual QD behaviors within the capability of current facilities.[23] After capping with a thin GaAs layer, the positions of quantum dots remain visible on the sample surface, which facilitate fabrication single QD device or experiment on a single QD.[24] In addition, the presented growth method can be further extended to obtain ordered QDs by combining the approach of fabricating ordered Ga droplets using focus ion beam or other techniques. [25]

All samples in this article were grown on semi-insulated GaAs (100) substrates using a molecular beam epitaxy (MBE) system equipped with reflection high-energy electron diffraction (RHEED). Substrate temperatures were measured by a noncontact transmission thermometry temperature measurement system and then calibrated by the native oxide desorption temperature of GaAs determined by RHEED. After heating the sample up to 610 °C, the sample was kept at this temperature for ten minutes to completely remove the native oxide. After oxide desorption, a GaAs buffer layer of 0.5 μm was grown at 580 °C. Then, the chamber was prepared for nanohole growth by Droplet Etching Epitaxy.[21, 26-29] The schematics of low density quantum dot growth are shown in Figure 1. First, all the cell shutters were closed, the As background flux pressure was reduced to ~ 10^{-9} torr and 3 ML Ga based on an equivalent amount of GaAs on (100) orientation was deposited to form droplets on GaAs surface as shown in Figure 1 (a). Subsequently, the sample was annealed at 600 °C for 5 minutes to form nanoholes. As shown in Figure 1 (b), the nanoholes are created by Ga nanodrilling under high temperature annealing. In this article, the substrate temperature (580 °C) was much higher than previous reports and thus the nanohole density was a lot lower. One reference sample A is grown with nanohole only and another three quantum dot samples were grown on nanohole templates. After the formation of nanoholes, the substrate temperature was immediately reduced

to 500 °C and different InAs coverages were deposited on the three samples at a growth rate 0.03 ML/s as illustrated in Figure 1 (c). The slow growth rate is essential for surface diffusion of In adatoms and nucleation of InAs QDs solely in nanoholes. The three quantum dot samples are assigned as samples B, C, and D, which correspond to InAs coverage of 0.9 ML, 1.2 ML, and 1.35 ML, respectively. The samples were then quenched to room temperature and then investigated by atomic force microscopy (AFM). A separated set of samples were grown by using the same recipes but the QDs were capped with 10 nm GaAs for photoluminescence (PL) study. Such a thin capping layer left distinct surface structures which can assist to locate the position of buried quantum dots, as shown in Figure 1 (d). These three samples are labeled as B', C', and D' corresponding to InAs coverages of 0.9 ML, 1.2 ML, and 1.35 ML, respectively.

Figure 2 shows the AFM images of the nanohole sample (sample A). Figure 2 (a) is a 20 × 20 μm^2 AFM image of the nanohole sample surface. The very low density of nanoholes is due to Droplet Epitaxy growth at the high temperature of 580 °C. The formation of nanodroplets is governed by two different mechanisms at different temperatures[22]. According to the classical nucleation theory, the density of clusters reduces with increasing temperature and the density of nanodroplets generally follows a classical nucleation scaling law at low temperatures. However, Ostwald ripening becomes substantial at high temperatures, and Ostwald ripening further reduce the density of droplets on GaAs surface.[30] Therefore, by growing at the high temperature of 580 °C, the obtained nanhole density of sample A is estimated as low as ~4×10^6 cm^{-2}. Figure 2 (b) shows a magnified 1 × 1 μm^2 AFM image of a single nanohole. The ring-like wall around the nanohole is from the droplet etching process.[20, 28, 29, 31] The presence of Ga droplet promotes the As removal from the substrate. As a result of low solubility around 10^{-4}, As atoms quickly diffuse through the liquid Ga droplet.[31] The released As atoms from the substrate reattach to the surface by reacting with Ga atoms diffused out from the liquid droplet and crystallize the ring-shaped wall at the boundary of the droplet.[32] The line profile shown in Figure 2 (c) is taken along [011] direction. The hole diameter is measured to be 147.8 ± 16.1 nm while the hole depth is observed to be as deep as 29.1 ± 3.5 nm. The hole depth value is significantly larger than previous reports.[20, 28] This can be attributed to enhanced As desorption from the

substrate at the high growth temperature. The etching depth is the product of etch rate, r_e, and etching time, t.[31] The etching rate, r_e, can be expressed as a thermally activated function, $r_e \propto \exp(-E_e / k_B T)$, where E_e is the activation energy of etching, i.e. As removal from the substrate, k_B is the Boltzmann constant, and T is temperature.[31] Therefore, the etching rate, r_e, can be significantly enhanced at high temperature due to the exponential dependence of temperature. As a result of enhanced nanodrilling by droplets at high temperatures, the nanohole depth is much deeper in this study. It has been known that the As desorption is the key process of the nanodrilling effect and As flux can be used to control the etching rate and the nanohole morphology.[33, 34] In order to create smaller nanoholes, a higher As flux or a shorter annealing time can be used.

Figure 3 shows 5 × 5 μm^2 AFM images of samples B, C, and D. As shown in Figure 3 (a-c), all samples show very low density nanoholes, of which the density is, similar as sample A, 5~7 × 10^6 cm^{-2}. Magnified AFM images of samples B, C, and D are displayed in Figure 3(d-f). The AFM images demonstrate the detailed structures of quantum dots formed in the nanoholes. It is also worth noting that the quantum dots only form inside the nanoholes, indicating the quantum dot nucleation took place preferably inside the nanoholes. The phenomenon can be understood by the modified surface chemical potential by Droplet Epitaxy. Specifically, the deposited atoms diffuse from regions of high chemical potential to regions of low chemical potential. Given a sufficient long diffusion length, this implies that the chemical potential in the nucleation sites is the minimum. To formulate the chemical potential quantitatively, we assume the InAs epitaxial layer conforms exactly to the shape of the underlying hole-patterned GaAs structures. The chemical potential for the epitaxial layer growth can be expressed as[35]

$$\mu = \mu_0 + \Omega\gamma k + \Omega E_{str} + \Omega \frac{\partial \gamma(h)}{\partial h} \quad (1)$$

where μ_0 is the thermodynamic driving force of the crystallization process, Ω is the atomic volume, γ is the surface energy density, k is the local surface curvature, and E_{str} is the mismatch strain energy density stored in the epitaxial material. The strain energy density can be expressed by

$E_{str} = 2G\left[(1+\upsilon)/(1-\upsilon)\right]\varepsilon_0^2$, where G is the shear modulus, υ is the Poisson ratio, and ε_0 is the strain mismatch between the epitaxial material and the substrate. The second term represents the contribution of the surface curvature, the third term determines the strain contribution to the chemical potential, and the last term is the contribution of the surface energy change with the increase of layer thickness. We use AFM-generated surface profiles in Fig. 2(c) to calculate the surface curvature and hence the locally varying surface chemical potential. The calculated result of the chemical potential of the hole-patterned GaAs substrate is shown in Fig. 4. The dashed curve is the surface profile of the hole-patterned surface, and the solid curve is the calculated surface chemical potential according to Eq. (1). One can find that there is a minima of the chemical potential at the center of the nanohole caused by the minimal surface curvature.

The position of the minimal surface chemical potential agrees very well with the observed locations of self-assembled InAs QDs inside the nanoholes, as shown in Fig. 3. The local minima of chemical potential at center of holes are narrow and relatively deep, which suggests that the possibility of QDs formed inside the nanoholes is high. By using a very slow growth rate of InAs, all QDs are formed in the nanoholes. The experimental observations are consistent with the modeling results based on the calculations of surface chemical potential. It should be noted that the anisotropies of the thermodynamic driving force, the surface energy density, and the mismatch strain energy density are not considered here.

The line profile taken along the [011] direction of all samples are shown in Fig. 3(h-j). The quantum dots preferentially form against the nanohole side wall along the [01-1] direction. Firstly, the nanohole formation is obviously an anisotropic process, which is evident from Figure 2. The nanohole is slightly deviated from perfect circle with a high GaAs hillock along [01-1] direction. In other words, the surrounding area of the nanoholes is higher along the [01-1] direction than that along [011] direction. We speculate that the anisotropic process during nanodrilling creates more atomic steps along the [01-1] side wall with a lower surface energy density, which results in lower chemical potential along the [01-1] direction preferred by the InAs quantum dot nucleation. By carefully control the droplet etching process, highly symmetric circular nanohole can be realized[36]. In addition, since the GaAs (100) surface is anisotropic, nanostructures

formed on the surface may deviated from symmetrical shape. It has been shown that the anisotropic strain field can lead to formation of slightly elongated InAs QDs along the [01-1] direction on GaAs surface[37]. Therefore, the anisotropic strain can also account for the preferred nucleation of InAs QDs against the sidewall along the [01-1] direction, according to the Equation (1). The anisotropic surface diffusion lengths can also lead to preferential nucleation of QDs along the [01-1] direction.

After InAs deposition, the nanoholes are only partially filled with QDs against the sidewall due to the anisotropic nucleation. The quantum dots of all samples show similar sizes following the shape of nanoholes. The measured average lateral size of these quantum dots is in the range of 50-80 nm, as shown in Fig. 5. The lateral width of the quantum dots measured along [01-1] is 65±10.8 nm, 55±10.8 nm, 74±.8.4 nm and the width measured along [011] is 71±13.7 nm, 60±9.4 nm, and 59±5.7 nm for samples B, C, and D, respectively. As shown in Fig. 5, the width of the QD of sample D is the largest along the [01-1] direction while the smallest in the [011] direction for all samples. It should be noted that due to the fact that the quantum dots are below the surface, the size measurement using AFM becomes less accurate. Therefore, the QD size measurement of sample B also has the largest error. Moreover, the height of the quantum dots is difficult to estimate as the quantum dots partially fill inside the nanoholes. Nonetheless, in Fig. 3, it shows that the tip of the quantum dot of sample B is well below the surface while the tip of the quantum dots of other samples almost reaches to the level of the surface. Obviously, the quantum dots grow taller with increasing In coverage and such an argument agrees with previous reports on QD fabrication with much higher In coverages.[20] To obtain QDs with smaller size, less In deposition can be applied during QD growth. At the same time, the nanoholes can also be grown with smaller dimensions by less annealing time and/or higher As flux. Therefore, the growth of QDs can be either independently controlled at the nucleation step or adjusted by designing the formation of nanoholes. As a result, the control of the QD density and QD size can be decoupled and optimized independently.

In order to gain more details of the QDs fabricated by nanohole filling, another set of QD samples with the same sample structures were grown but capped with a 10 nm GaAs cap layer at 500 °C after the

formation of InAs QDs. The AFM images of the capped samples B', C', and D' are illustrated in Figure 6. After being caped with the thin GaAs layer, the QD position is still clearly present on the surface. This is so because the nanohole surrounding structures are made of GaAs and subsequent growth of the thin GaAs layer tends follow the original structure shape. The surface can be flattened by further deposition of GaAs. As previously discussed and shown in Fig. 4, the nanohole region has lower chemical potential, and nucleation of GaAs also preferentially happens in the nanoholes. For samples B', C', and D', the depth of the hole left on the surface is only few nanometers after GaAs capping. It is also worth mentioning that the unclosed hole depth decreases with increasing In coverage. The hole depth of samples B' and C' is ~2.5 nm while the hole is totally filled for sample D'. This again indicates that the QD size is dependent on the In coverage.

Photoluminescence measurements of these caped low density InAs QDs were carried out at 77 K by using a Fourier transform infrared spectrometer. The excitation power intensity is estimated to be about ~1.0 W/cm^2. Figure 6 (d) shows the PL spectra of all the capped samples. The PL spectra for samples B', C', and D' are normalized with the peak intensity. All the three samples exhibit a single narrow emission peak. As shown in Figure 6 (d), samples B', C', and D' show PL peaks at 0.93 μm, 0.96 μm, and 1.0 μm with full width at half maxima (FWHM) of 14 nm (20 meV), 17 nm (23 meV), and 26 nm (33 meV), respectively. The narrow PL peak width indicates formation of uniform low density QDs in the nanoholes. The red-shift of PL peak upon increasing In coverage suggests weaker quantum confinement of QDs with higher In coverage and thus, formation of larger QDs. Therefore, the PL results again confirm that the size of QDs increases from inceasing In coverage from 0.9 ML to 1.35 ML. It is interesting to observe that the FWHM grows much larger for sample D' compared with that of sample B'. With increasing In coverage, the QDs grow larger and expand out from the holes, resulting in a large sizer variation and hence larger inhomogeneity broadening of PL. When the In coverage is low, the QDs are confined inside the nanoholes. When the In coverage increases, the QDs grow bigger and the overall uniformity gets worse. Therefore,

FWHM increases with increasing In coverage. Nevertheless, the narrow and well-resolved PL spectra of such low density QDs at 77 K indicate good optical property and uniformity of these QDs.

To gain further insights into the optical properties of the QDs, power dependent PL measurements of the QDs were investigated at 15 K. The power dependent PL measurements are performed using the 532 nm line of a doubled neodymium-doped yttrium aluminum garnet (YAG) laser for excitation in variable temperatures, 10–300 K, within a closed cycle helium cryostat. The laser spot diameter is about 20 μm. The excitation power intensity is varying from I_0=70 mW/cm^2 to 2.4×10^4 I_0. The PL signal from the sample is dispersed by a 0.5 m single-grating monochromator and detected by a LN-cooled InGaAs photodiode detector array. Figure 7 (a)-(c) shows the power-dependent PL spectra for the three capped QD samples B', C' and D'. The PL spectral shape shows strong dependence on the excitation power. The PL peaks around 1.5 eV are due to GaAs emission from the substrate. At a low excitation power, the three samples exhibit a single narrow emission peak similar to what has been measured for low power excitation at 77 K. With increasing excitation power over 200 I_0, the PL spectra clearly show enhanced broadening. The broadening of PL emission mainly extended to shorter wavelength, which can be attributed to multiple additional transitions starting to appear at shorter wavelength range with the high excitation intensity. Due to state-filling, radiative recombination from excited states become substantial in the quantum dots, leading to broad emission spectra. As the QDs are relatively large in size, a weak quantum confinement with a large number of closely-spaced excited states is expected. As a result, the state-filling does not show well-resolved emission peaks from excited states. Additionally, a distinct narrow peak is observed below the GaAs bandgap around 833 nm – 845 nm at high laser excitation powers. The narrow emission peak is in the typical wetting layer spectral range of InAs/GaAs samples, which indicates formation of InAs wetting layer in addition to InAs quantum dots.[26] Based on the QD volume and density, it suggests that a portion of the InAs deposited on the surface does not contribute to the formation of QDs, and consequently, should deposit between nanoholes, hence forming a wetting layer.

In conclusion, ultralow density InAs quantum dots are grown on a self-assembled low density nanohole template patterned by high temperature Droplet Etching Epitaxy. Atomic force microscopy and photoluminescence technique have revealed that the quantum dot size can be controlled by varying the In coverage. The formed surface structure after capping quantum dots with a thin GaAs layer serves as the nanostructure position marker which benefits single QD device fabrication. The density and the size of the QDs can be controlled independently by using the method reported in this article. The presented low density nanostructure growth technique opens opportunities for studying single QD properties and single QD devices.

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Figure Captions

Figure 1. Schematics of low density InAs quantum dots growth on nanohole template. (a) Ga deposition in As depleted ambience to form low density Ga droplets by high temperature droplet epitaxy. (b) High temperature annealing to form nanholes. (c) Deposition InAs at a very low growth rate to create quantum dots in nanoholes. (d) Cover InAs quantum dots with a thin GaAs capping layer.

Figure 2. (a) A 20 × 20 μm^2 AFM image of sample A with nanoholes. (b) Magnified AFM image of a single nanhole. (c) Line profile of the nanohole in (b) along [011] direction.

Figure 3. (a-c) AFM images with lateral scale 5 × 5 μm^2 of sample B (0.9 ML InAs), sample C (1.2 ML InAs), and sample D (1.35 ML InAs). (d-f) Single QD AFM images of samples B, C, and D. (g-j) AFM line profile of sample B, C, and D taken along [011] direction. The lateral scale of the AFM images is 1 × 1 μm^2.

Figure 4. Chemical potential as a function of lateral distance of hole-patterned GaAs substrate. The thickness of InAs layer is 1.35ML in the calculation. The dashed blue line is the surface profile measured by AFM, and the black solid line represents surface chemical potential.

Figure 5. The lateral dimensions of the quantum dots formed in the nanoholes. The measurements are taken along [011] and [0-11] directions.

Figure 6. 5 × 5 μm^2 AFM images of samples (a) B', (b) C', and (c) D' after capping with 10 nm GaAs. (d) Photoluminescence of samples B', C', and D' measured at 77 K.

Figure 7. Photoluminescence spectra of samples (a) B', (b) C', and (c) D' measured with different excitation powers at 15 K. The excitation powers used are I_0=70 mW/cm^2, 20 I_0, 200 I_0, 3500 I_0, 2×10^4 I_0, and 2.4×10^4 I_0. The arrow in the plot indicates the direction of increasing laser excitation power.