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Controlled fabrication of InGaAs quantum dots by selective area epitaxy MOCVD growth

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Abstract

Control over the location, distribution, and size of quantum dots is essential for the engineering of next-generation semiconductor devices employing these remarkable nanostructures. We describe two approaches for achieving some level of this control in the InGaAs/GaAs material system. The first allows a degree of spatial selectivity by using strain differences in patterned InGaAs thin films as preferential sites for quantum dot growth. This method results in patterns of dots similar to those grown by self-assembly on an unpatterned InGaAs layer. The second method employs more conventional selective area epitaxy using a thin silicon dioxide mask patterned by electron beam lithography. This method allows control over the location of each quantum dot and variation of dot size through manipulation of the mask pattern. We present data on arrays of highly uniform InGaAs quantum dots fabricated in this manner.

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1. Introduction

Semiconductor lasers employing quantum dot active layers [1,2] grown by both metal-organic chemical vapor deposition (MOCVD) [3–6] and molecular beam epitaxy (MBE) [7–10] have been demonstrated to have advantages over lasers based on quantum well active layers, specifically in the areas of reduced threshold, beam quality [11], and operating temperature insensitivity [12]. Additionally, single electron switching devices can be used to form extremely dense computational systems with ultra low power consumption [13]. The application of quantum scale nanostructures to both optoelectronic and electronic integrated devices depends on the ability to engineer the parameters of the structures. These parameters include the size, size distribution, and material composition as well as their spatial

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location. Self-assembly has been the primary means for fabrication of large area arrays of quantum dots, and confinement of the region of self-assembly has provided limited spatial control of quantum dot growth [14–16]. Various techniques have been employed to explicitly define quantum dot nucleation sites and gain control over their emission properties. These include in situ electron beam lithographic patterning and selective area growth by MOCVD [17] and MBE [18,19], MBE growth on SiO_2 patterned by interferometric lithographic techniques [20], and electron beam patterning and etching of previously grown quantum well structures [21]. We report two methods for the controlled growth of quantum dots. The first uses pre-existing InGaAs thin films grown by selective area epitaxy to encourage quantum dot formation in localized regions. The quantum dot physical parameters can be varied through manipulation of the local InGaAs film thickness. We have demonstrated the area-controlled growth of quantum dots in controlled regions using this method. The second method uses electron beam lithography coupled with selective area epitaxy MOVCD [22–24] to explicitly define the dots' physical parameters and growth location on an individual basis. This technique is advantageous as it leaves the crystal surface unaffected and promotes the growth of coherent, dislocation free dots. We have demonstrated the fabrication of highly uniform, patterned arrays of quantum dots with a narrow emission linewidth using this method.

2. Area-controlled growth using InGaAs thin films

Area-controlled growth of quantum dots using InGaAs films as preferential dot growth locations has been demonstrated. Initially, a thin (1–2 monolayer) 15–20% InGaAs film is grown by selective area epitaxy using a SiO_2 growth inhibition mask on a GaAs (1 0 0) substrate. Growth rate enhancement effects may be used to modify the thickness of the film in different regions. The SiO_2 mask is then stripped and the sample is returned to the reactor for growth of the quantum dots. 0.5–1.5 monolayers of 50% InGaAs are then deposited between 500 and 550 °C. This deposition is intentionally below the critical coverage for quantum dot formation on a uniform GaAs substrate. The pre-existence of InGaAs in localized regions allows critical coverage to be obtained in those regions, while elsewhere, subcritical deposition occurs. The result is the growth of self-assembled quantum dots in the InGaAs patterned regions and 2D islands on the GaAs regions. Quantum dot densities of $4 \times 10^{10}\,cm^{-2}$ were achieved on the InGaAs thin film. Fig. 1 contains AFM scans taken of the surface of an InGaAs ridge and the GaAs region surrounding the ridge after the regrowth of quantum dots. Quantum dots are observed on the InGaAs ridge while a smooth, dot free region is observed in the region without InGaAs. It should be additionally noted that, unlike traditional selective area growth using a SiO_2 growth inhibition mask, no growth rate enhancement is observed since uniform deposition is allowed to occur everywhere on the sample. The result is uniformly sized quantum dots over the entire stripe regardless of its lateral size and geometry. Preferential quantum dot nucleation occurs only as a function of the InGaAs film thickness.

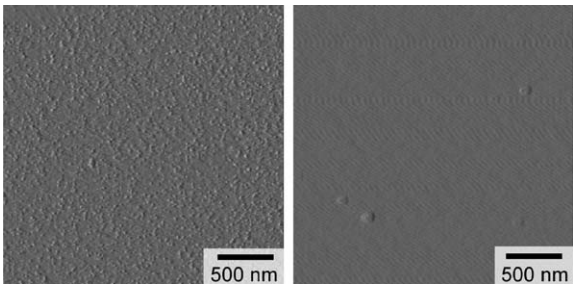

Fig. 1. AFM images of dot formation on an InGaAs ridge (left) and planar growth on the GaAs substrate (right).

3. Site-controlled quantum dot growth by selective area epitaxy

Site-controlled growth of InGaAs quantum dots, using electron beam lithography and selective area epitaxy to explicitly define the location and physical dimension of each individual quantum

dot, has been demonstrated. Initially a base structure was grown on an n+ GaAs substrate which consists of an n-type, 1 μm thick, 74% AlGaAs layer followed by an intrinsic, 100 nm thick, GaAs layer. The substrate was then removed from the reactor and 100 Å of SiO_2 was deposited by plasma-enhanced chemical vapor deposition (PECVD). The oxide was patterned by conventional optical lithography using a "template" pattern consisting of 450 μm stripes. The SiO_2 is then etched with a dilute (4:50) buffered hydrofluoric acid solution. As in conventional selective area epitaxy, a large fraction of the sample must be exposed during the regrowth or enhancement effects result in an uncontrollably high growth rate. The purpose of the template pattern is to remove the oxide mask over the bulk of the sample, leaving oxide only in the regions that will be patterned via electron beam lithography. The template eliminates the need to write the field of the sample during electron beam lithography, significantly reducing write time. The substrate was then patterned again via electron beam lithography in 3% 950 K polymethylmethacrylate (PMMA) to create the quantum dot growth mask. The electron beam pattern consists of 45 nm holes arranged on a triangular lattice with center to center spacings of 125, 100, and 75 nm corresponding to dot densities of 7.4×10^9, 1.2×10^{10}, and $2.1 \times 10^{10}\,cm^{-2}$, respectively. Typically an area approximately 200 μm wide and 1–2 mm long is written for each pattern. A scanning electron microscope image of the developed electron beam resist pattern is shown in Fig. 2. The pattern was etched into the oxide with a dilute (4:50) buffered hydrofluoric acid solution. While reactive ion etching would seem a more appropriate means of transferring the pattern to the oxide, it introduces too much damage to the semiconductor surface to allow the regrowth of coherent, dislocation free material. Wet etching avoids this problem, leaving a clean, damage free crystal surface, but it does result in significant enlargement (20–30 nm) of the diameter of the quantum dot growth mask features.

After thorough cleaning, the substrate is returned to the reactor for the growth of the quantum dots. 50% InGaAs dots followed by a thin layer (~1 nm) of GaAs were grown at 600 °C. The deposition time was determined experimentally to produce quantum dots approximately 100 Å high for source flows corresponding to a 1:1 TMG:TMI gas phase composition. The sample was then removed from the reactor and the oxide growth mask was removed with buffered hydrofluoric acid. Fig. 3 contains an atomic force microscope rendering of the 75 nm period structure after growth. The growth rate enhancement

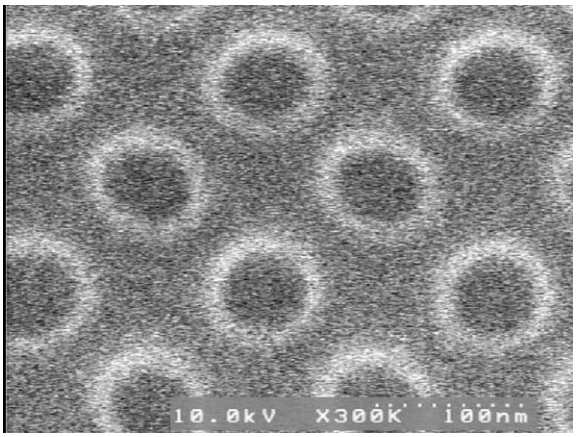

Fig. 2. SEM image of lithographic pattern used as template for growth of patterned quantum dots.

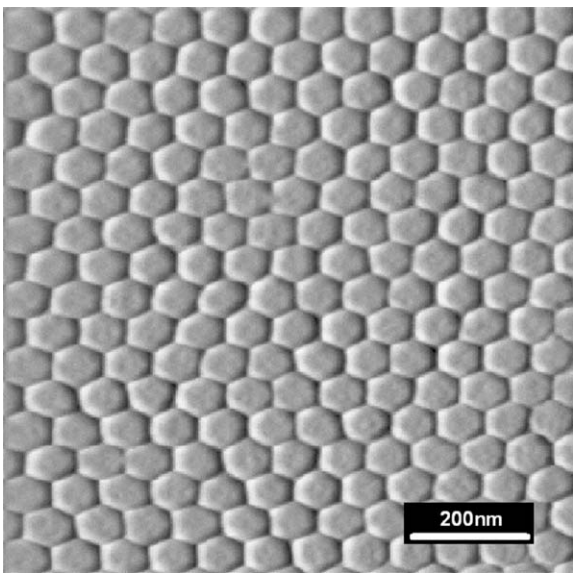

Fig. 3. AFM rendering of InGaAs quantum dot structure after growth.

factor is a function of both the deposition parameters (flow rates and time) and the lateral dimensions of the growth mask. The latter of these effects can be described as two simultaneous enhancement effects. First, there is a conventional growth rate enhancement effect caused by lateral diffusion of growth precursors into the dot growth region resulting from the SiO_2 *surrounding* the patterned region. This effect extends several micrometers in from the edges of the patterned area. It results in a growth rate enhancement factor which decays with distance from the bulk oxide surrounding the pattern and increased growth rate enhancement around the periphery of the patterned region. Since the ultimate goal is highly uniform quantum dot arrays, this is an undesirable effect. This problem can be avoided by simply writing the pattern larger than necessary such that the larger dots are formed outside of the device active region. The second enhancement effect results in a uniform growth rate enhancement within the patterned region. The quantum dot growth mask features are very small compared to the gas phase diffusion length of the growth precursors so lateral diffusion is irrelevant. The partial coverage of the semiconductor surface by the oxide mask results in a localized precursor concentration enhancement and, thus, a simple areal growth rate enhancement. The value of the enhancement is related to the fill factor of the template. As long as the enhancement factor is not too high so as to result in uncontrollable growth rates, this effect is not detrimental to the fabrication of highly uniform dots. Since we desire a high density of dots, the oxide fill factor will always be low enough to produce a manageable growth rate enhancement.

The sample was then returned to the reactor for the third and final growth step. A 100 Å, 20% InGaAs QW was grown at 550 °C followed by 100 nm of intrinsic GaAs, 1 μm of p-type, 74% AlGaAs and 100 nm of p+ GaAs at 600 °C. The growth was started immediately after a temperature ramp from room temperature to 550 °C in order to minimize any surface reformation of the quantum dots. The sample was then fabricated into broad area diodes. First, the p+ contact layer was patterned into 150-μm stripes centered over the patterned quantum dot regions by optical lithography. The wafer was thinned to facilitate cleaving and metal contacts were deposited on both sides. Several devices where dots had not been patterned on the same sample were processed as well to act as controls. These devices contain only the quantum well used to cap the dots as an active layer. The sample was then cleaved into individual diodes 2 mm in length and tested under pulsed current conditions.

4. Emission studies on site-controlled patterned quantum dots

Room temperature electroluminescence (EL) was observed from the diodes under 500 mA of pulsed current with a duty cycle of 2% and 2-μs pulses. The emission spectrum from the quantum dot devices is shown in Fig. 4. The quantum dot emission peak is centered at 1080 nm and is 55 meV wide at the half maximum point. The wavelength is considerably shorter than expected from quantum dots of this size and composition. We believe the significant blue shift results from indium desorption and interdiffusion before and during, respectively, the growth of the upper core and cladding. The linewidth is comparable to that of the best reported room temperature EL from

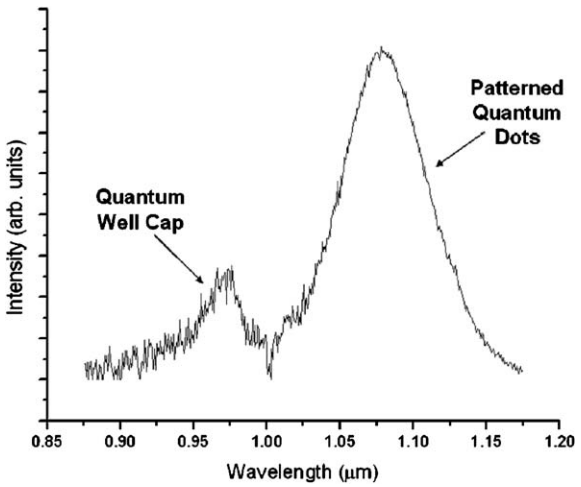

Fig. 4. Electroluminescence spectra of diodes containing patterned quantum dots with quantum well cap.

self-assembled quantum dots grown by MBE (55 meV) [25]. It is slightly wider than the best reported room temperature EL from MOCVD grown self-assembled dots (40 meV) [26], but was observable at much lower current densities (170 A/cm^2 vs. more than 330 A/cm^2). The diodes containing only a QW demonstrate an emission spectrum centered at 970 nm with a 49 meV full-width at half-maximum. The narrow emission linewidth of the quantum dot devices implies a narrow size distribution for the patterned quantum dot active layer. The weak relative intensity of the quantum well emission peak on the quantum dot device spectrum suggests that strong coupling between the dots and the well has been achieved. These results indicate that the fabrication process detailed above is compatible with the production of optical quality, spatially controllable nanostructures for use as a gain medium in optoelectronic devices. We have not yet achieved laser action from these devices most likely because of crystal quality degradation in the layers immediately above the active region. Efforts are under way to optimize the quantum dot overgrowth process and resolve these problems.

5. Conclusion

We have demonstrated two methods for controlling the location and physical parameters of quantum dots in the InGaAs/GaAs material system. The first allows for the large area growth of self-assembled quantum dots in selected areas and utilizes prepatterned InGaAs thin films as the control mechanism. This mechanism is ideal for integrated photonic devices utilizing large areas of quantum dots as active layers since multiple dot regions with varying parameters can be grown simultaneously. The second method uses electron beam lithography and direct selective area epitaxy to explicitly define the location and physical parameters of each individual quantum dot. We have demonstrated highly uniform arrays of optical quality InGaAs quantum dots on a GaAs substrate using this growth technique. Implicit in this technique is the ability to control the size, location, and area distribution of quantum dot active regions which provides a large degree of engineering freedom to both localize the dots and control their emission wavelength. Diodes fabricated from these structures have demonstrated emission spectra centered near 1080 nm with full width at half maximum values of 55 meV, which is comparable to reported MBE and MOCVD grown self-assembled quantum dots.

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