

In as Quantum Dot Self-Assembly via Interference Lithography: Implications for Nanomedicine and Diagnostic Technologies

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ABSTRACT

Laser interference lithography is used to directly pattern the growing surface during molecular beam epitaxy growth of self-assembled InAs quantum dots on GaAs (100) substrates. Arrays of few-monolayer high nano-islands are formed prior to InAs quantum dot growth, which we believe result from the surface diffusion promoted by transient photothermal gradients. The deposition of InAs on such a surface leads to the nucleation of quantum dots solely at the island sites. The number of dots per site is determined by the island size which varies with the laser energy intensity. We are able to achieve highly ordered dense arrays of quantum dots with a single nanosecond laser pulse exposure. InAs quantum dots formed in this fashion show bright narrow photoluminescence with a peak at 1.04 eV at 88 K.

Keywords: InAs quantum dots, semiconductors, interference lithography, nanopatterning, surface diffusion, photoluminescence.

1. INTRODUCTION

Semiconductor InAs quantum dots (QDs) have become the subject of intense research due to their quantum properties, which make them important building blocks for quantum information processing and computation [1,2]. Epitaxial self-assembly methods have emerged in which a morphological instability on the growth surface induces the formation of small defect-free semiconductor QDs. By careful choice of growth conditions control over the dimensions can be achieved to nm precision, resulting in structures which are beyond the limitations of present-day conventional photolithography. However, epitaxial self-assembly has a major limitation in that atoms migrate randomly on a surface until they eventually coalesce into stable islands. As a result, unstructured self-assembly leads to stochastic distribution of sites and an inhomogeneous distribution of sizes. This will be a major block to device integration. Precise positioning in arrays of identical QDs will be critical for future device implementation.

As a response, directed (or templated) self-assembly methods have been studied which seek to use lithographic techniques such as electron beam lithography (EBL) or focused ion beam lithography (FIB), in combination with surface etching [3,4]. The typical approach is to create nanohole hole-templated substrates using ex-situ processing methods and then introduce these to deposition equipment with which we perform epitaxial re-growth. Epitaxial re-growth methods are of course well established, but it is important to note that the scale required to nucleate individual atoms is several orders of magnitude smaller than for conventional device applications. As a result, considerable effort is required to clean the surface and to prepare this to create sites for nucleation on the atomic scale. The methods are demanding and complex and the combination of ex-situ processing and in-situ growth leads to a low throughput which will not lend itself easily to scaling up to device production. In situ methods, compatible with the epitaxial nucleation step, would be a highly attractive alternative. Laser interference lithography (LIL) has emerged as a possible route for in situ surface patterning in recent years, offering rapid and large area of the semiconductor surface. As a non-invasive technique, this can be applied to epitaxial reactors with appropriate optical access to the substrate. In-situ interference was first reported by Clegg et al who applied a two-beam pattern to an InGaAs layer grown by molecular beam epitaxy (MBE) to form a modulated pattern of QDs [5]. The results were interpreted as due to indium atom migration from the interference maxima to minima regions, where they then nucleate to form QDs. However, this result has a limited application in that it only results in a weak modulation of the QD density rather than specific site location.

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Subsequently, Zhang et al have reported the use of 4-beam interference to grow QD arrays [6]. The results show good site control, but with a large variation in the QD number per site and a broad distribution of dot sizes. The authors attribute their observations to indium desorption at the interference maxima rather than diffusion, which contradicts the observations of Clegg. In this work, we report the self-assembly of InAs QD arrays directed by four-beam laser interference based on a surface diffusion mechanism. The realized QD structures show precisely organized arrays with improved QD statistics and narrow photoluminescence (PL) results.

2. EXPERIMENTAL DETAILS

Samples were grown on 2 inch GaAs (100) substrates using conventional MBE. After oxide desorption, a 500 nm GaAs buffer layer was grown at a growth rate of 1 ML s^{-1} and a substrate temperature of $T = 600^\circ\text{C}$. The substrate temperature was then decreased to 500°C for the growth of InAs. 1-1.5 monolayers (ML) of InAs were deposited at a low growth rate of 0.026 ML s^{-1} . During the InAs growth process, in-situ single pulse interference irradiation was performed after 1 ML InAs deposition, which is below the critical thickness for 2-D to 3-D transition. The laser pulse is *P*-polarized operating at 355 nm from a flashlamp pumped Nd:YAG laser (Innolas Spitlight), with a pulse width of 7 ns, a beam diameter of 5 mm and pulse energy of 40 mJ. The simplified schematic diagram of MBE-LIL system is demonstrated in Figure 1. Four coherent laser beams are introduced to the substrate surface at an angle of incidence of 58° and azimuth angles of 0° , 90° , 180° , 270° via four symmetric anti-reflective optical vacuum viewports facing upwards and positioned in the lower half of the MBE system.

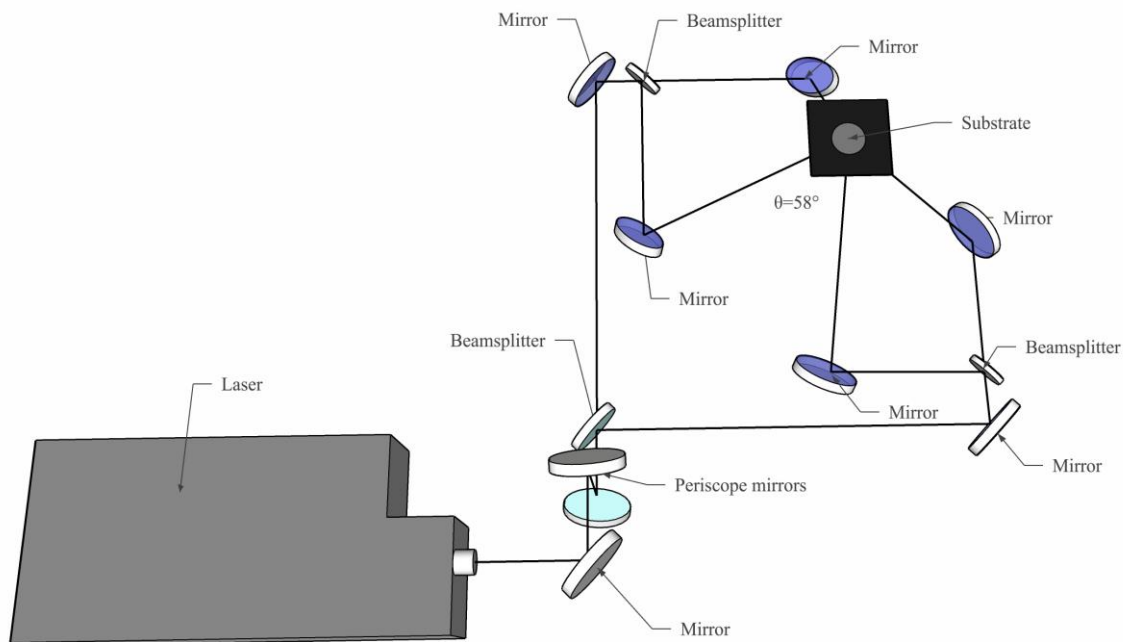
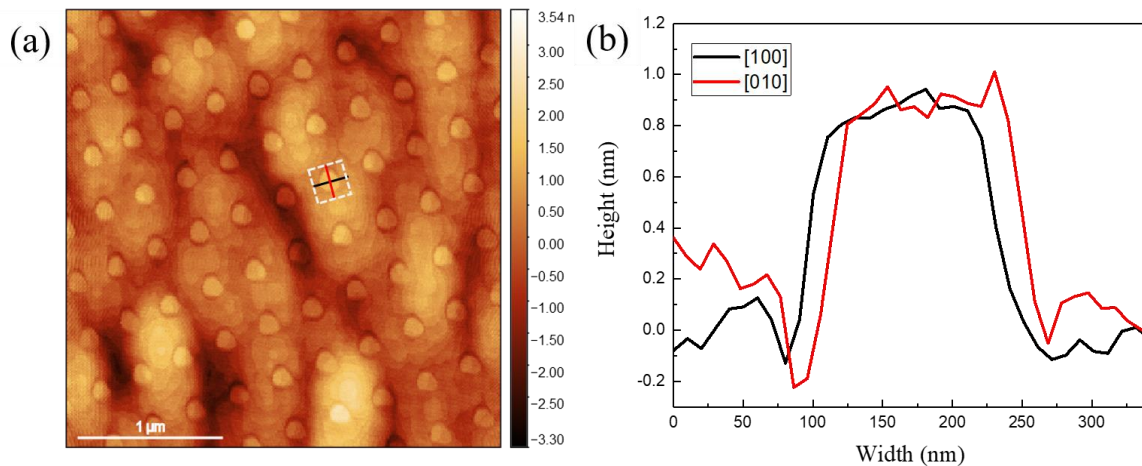


Figure 1. Schematic diagram of MBE-LIL configuration.

3. RESULTS AND DISCUSSION

Figure 2 shows an atomic force microscope (AFM) image of an uncapped 1 ML InAs layer surface that patterned by laser interference with a square array of nano-islands spaced 300 nm apart and the cross-sectional profile of an individual nano-island. The nano-islands observed are generally 80-150 nm wide and 0.5-1 nm (2-3 ML) high. The formation of these shallow islands is associated with lower pulse energies in the range of $10\text{-}30 \text{ mJ/cm}^2$. Due to the absorption of the laser pulse energy, heat is generated in the near surface. From our previous calculations [7], the surface temperature rises very

rapidly after the pulse with the highest temperature located in the interference maxima. Therefore, a transient thermal gradient is created on the surface. Consistent with the methodology proposed by Clegg et al [5], we attribute the nano-island formation to thermal-induced surface diffusion process. The results are in accordance with a model of indium surface diffusion under the influence of a transient thermal gradients, where the indium atoms diffuse from the 'hot' regions



(interference maxima) to 'cold' regions (interference minima) [7]. Accumulation of indium in the 'cold' regions then forms patterned nano-islands.

Figure 2. (a) A $3 \times 3 \mu\text{m}^2$ AFM image of a sample surface with 1 ML InAs deposition after LIL. (b) Line profiles through the nano-island along the [100] and [010] directions marked in (a).

For the following samples, additional 0.5 ML of InAs were deposited after the pulse on the 1 ML InAs surface at the same growth rate. For the growth on such an island-structured surface, we would expect that the nucleation of QDs to be strongly influenced by the surface morphology. This is indeed what we observe; InAs QDs are preferentially nucleated at the edge of islands (see Figure 3(a)), and as the size of island reduces, more QDs are formed (Figure 3(b)). The nucleation of QDs at the edge of the nano-islands is highly reliable and has been observed in more than 20 samples. Corresponding three-dimensional AFM images are displayed in Figure (c-d). The size variation of nano-islands results from an uneven laser energy distribution. The laser beam we used has a Gaussian intensity profile, therefore, we always found that smaller islands are formed away from the centre area, where the energy is relatively low. At the limits of the smallest island size, we find the island is almost consumed and we only observe ordered arrays of InAs QDs with the same pitch as the nano-islands. Figure 3(e) presents an AFM image of such QD arrays directed by interference lithography. The patterned QDs typically have a width of 40-50 nm and height of 12-15 nm. The QD density is set by the interference pitch and is relatively low ($\sim 1.5 \times 10^9 \text{ cm}^{-2}$). For comparison, the corresponding non-patterned region (edge of sample) shown in Figure 3(f) has a high density ($\sim 1 \times 10^{10} \text{ cm}^{-2}$) of small QDs and quasi-3D islands. The results indicate that the InAs coverage is at or just below the critical thickness for the growth of QDs [8]. Returning to the patterned regions, the results suggest that under the influence of the laser pulse, indium atom diffusion takes place from the interference maxima regions to minima regions. With a decreased size of nano-islands, more In atoms can accumulate within the locally concentrated area, giving rise to a high nucleation rate for QDs. Consequently, in the interference minima regions, the InAs coverage can easily reach the critical thickness for 2D-3D transition while such growth is largely suppressed on the planar regions between the patterned sites or indeed on the non-patterned regions at the edge or the wafer. The InAs QDs formed in this way present enlarged dot size and better size uniformity. The number of QDs per site is mainly dependent upon the island size resulting from the locally varying laser energy intensity.

Samples for PL characterization were capped with a 200 nm GaAs layer. In order to improve the PL signal, a 300 nm layer of AlGaAs is grown prior to GaAs buffer layer. The low-temperature PL measurements were performed ex situ with the sample mounted in a continuous flow cryostat cooled to around 88 K by liquid nitrogen. A 659 nm continuous-wave pump laser was focused through a $20\times$ objective lens to a light spot with a radius of 5 μm . Figure 4 presents a PL spectrum of patterned InAs QDs excited by the laser power of 10 μW . A strong PL signal at the ground state is observed with a peak wavelength of 1193 nm. A narrow linewidth of 22 meV is observed, which is comparable with some of the best reports in

the literature. The results indicate that the application of laser interference lithography does not degrade the quality of QDs, but that indeed these show a high optical quality. Further optimization is however required, particularly in terms of

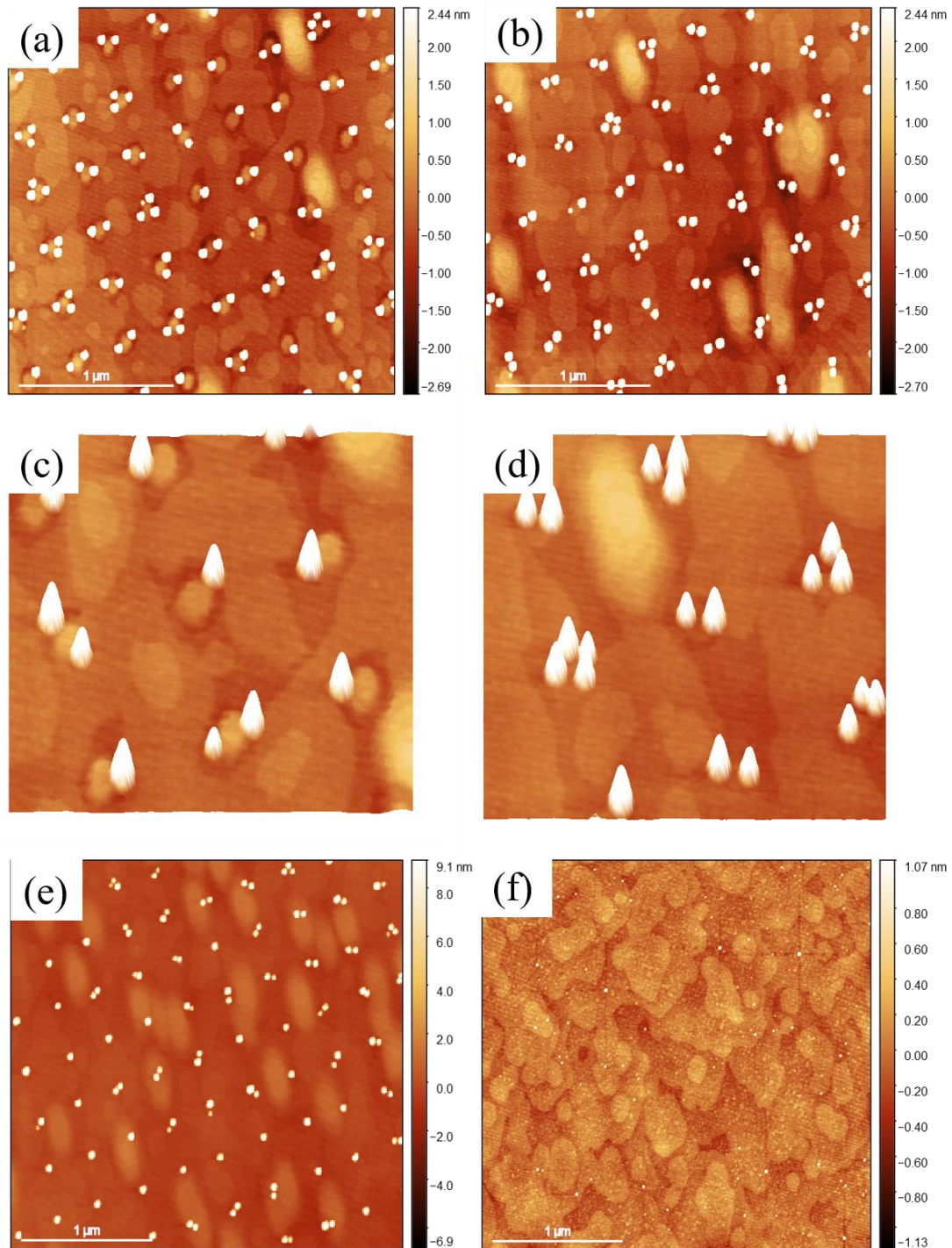


Figure 3. (a) A $2.5 \times 2.5 \mu\text{m}^2$ AFM image of a sample surface of InAs QD nucleate at the edge of big nano-islands. (b) QD nucleate at the edge of small nano-islands. (c-d) $1 \times 1 \mu\text{m}^2$ three-dimensional AFM images corresponding to (a-b). (e) A $3 \times 3 \mu\text{m}^2$ AFM image of InAs QD array with a pitch of 300 nm. (f) Non-patterned region of QDs in the same sample.

site occupancy. We also need to examine the single dot emission statistics, which is proving a challenge because the relatively small pitch results in a large number of dots ($\sim 10 \mu\text{m}^{-1}$) within the diffraction limited micro-PL spot size.

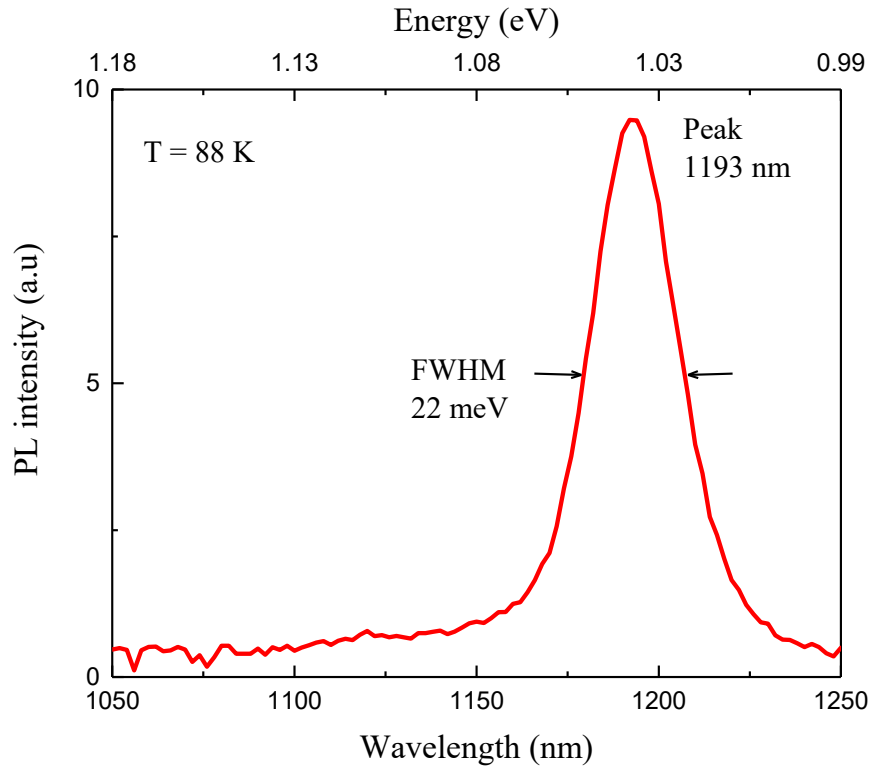


Figure 4. PL spectrum of the interference-induced InAs QD ensembles excited at the temperature of 88 K and excitation laser power of 10 μ W

In our work, we suggest that the formation of nano-islands is the precursor for subsequent QD formation that QDs are nucleated at small islands (<100nm) in which the local InAs coverage reaches the critical thickness. In a previous study by Zhang et al [6], there is no correlation between the QD pattern and the underlying morphology and it seems difficult to explain the formation of nano-islands solely through desorption of large areas of the surface. Consistent with Clegg et al [5], we observe strong justification for a diffusion-based mechanism with a well-defined temperature threshold and with rapid diffusion over the short timescale of the laser transient which creates a patterned morphology which can act as a nucleation site for QDs and perhaps for other nanostructures.

4. CONCLUSION

In conclusion, we demonstrate the growth of optical characterization of InAs QDs on GaAs (100) substrates. In situ laser interference lithography is applied in a molecular beam epitaxy chamber to directly pattern the growing surface. The self-assembly is guided through the diffusive kinetics under thermal gradients to achieve the lateral ordering of QDs. Square arrays of nano-islands with a pitch of 300 nm provide the energetically favourable nucleation sites for QDs where the dots prefer to nucleate at the edge of islands. Good ordering of single QDs or QD pairs has been obtained in the presence of nano-islands, and exhibit a good optical quality from the photoluminescence characterization. This technique offers a fast and efficient way to achieve highly-ordered nanostructures for the realization of novel photonic devices or quantum information technologies.

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