Precise Arrays of Epitaxial Quantum Dots Nucleated by In-Situ Laser Interference for Quantum Information Technology Applications

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ABSTRACT: Precisely ordered arrays of InAs quantum dots are formed on a nanoisland-structured GaAs (100) surface using in-situ laser interference during self-assembled molecular beam epitaxial growth. Nanoislands induced by single-pulse four-beam laser interference act as preferential nucleation sites for InAs quantum dots and result in site occupation dependent on the size of nanoislands, the InAs coverage and the laser parameters. By optimizing the growth and interference conditions, regular dense ordering of single dots was obtained for the first time using this in-situ noninvasive approach. The photoluminescence spectra of the resulting quantum dot arrays with a period of 300 nm show good optical quality and uniformity. This technique paves the way for the rapid large-scale fabrication of arrays of single dots to enable quantum information technology device platforms.

KEYWORDS: Quantum dots, nanostructures, InAs, nanopatterning, laser interference, III-V compound semiconductors

INTRODUCTION

Low dimensional semiconductor nanostructures such as quantum dots (QDs) and nanowires (NWs) have shown significant potential for applications in optoelectronics and nanoelectronics, e.g., low-threshold lasers, single electron transistors and quantum memory devices,1-3 owing to the unique nature of their quantized electronic states. III-V QDs obtained by self-assembly via the Stranski-Krastanov (S-K) growth mode exhibit high structural and optical quality. As ideal solid-state single photon or entangled photon sources, self-assembled QDs have enabled a wealth of new physics and device applications in the fields of quantum information processing and communication.5,6 For instance, single QDs coupled to a microcavity mode are able to achieve long-range spin-spin interactions7 and long spin decoherence times8 for electrons and holes were reported based on spin-based qubits in III-V semiconductor QDs.9 The strong coupling between excitonic and photonic states enables the manipulation of quantum information.4 However, most of this work to date has been performed on isolated QDs, usually by carefully selecting an individual dot from the ensemble, which is time-consuming and results in a very low yield when integrating single QDs into photonic cavities or other quantum circuits. Since unstructured QD epitaxial growth has size and site inhomogeneity as a consequence of a stochastic nucleation process,10 it would be difficult to incorporate such single QDs or QD molecules into device arrays. In addition, densely ordered QDs show the possibility of long-range quantum correlations, which is promising for quantum computation.11 It has also been suggested that the size distribution of QDs can be improved by lateral ordering the dots.12 Therefore, the precise positioning in arrays of identical QDs is highly desirable for future quantum information technology device applications.

As a response to these issues, strategies to achieve lateral alignment of QDs have been extensively explored, in which lithographic techniques such as electron beam lithography (EBL)13 and focused ion beam lithography (FIB)14 have been used to create hole-templated substrates prior to QD growth. However, despite some excellent achievements15,16 these are low throughput methods and do not lend themselves easily to device production. In addition, as existing techniques, defects and impurities on the atomic scale are introduced due to the heterogeneous processing and this limits performance. In-situ methods, compatible with the epitaxial nucleation step, would be a highly attractive alternative. Interference lithography has emerged as a possible route for in situ surface patterning in recent years, offering rapid and large area structuring of the semiconductor surface. As a non-invasive technique, this can be applied to epitaxial reactors providing there is appropriate optical access to the substrate. Semiconductor nanostructures recently reported using this process include planar nanowires guided by two-beam interference17 and the formation of QD arrays by two-beam interference.18 The significance these works is that they show that near surface absorption of short (few ns) pulses of moderate energy (~50 mJ) is sufficient to induce surface nanostructuring during III-V epitaxy. However, the underlying mechanisms behind these observations are somewhat unclear, with Clegg et al.19 suggesting a surface diffusion process while Zhang et al.19 attributing their observations to the thermal desorption of indium. None of these shows the presence of an intermediate surface structuring step before the nucleation of the nanowires or QDs. Furthermore, precisely controlled single dot arrays were not achieved and associated optical properties have never been reported.

In this work, we have investigated the growth, structural and optical properties of InAs QD arrays formed in-situ during molecular beam epitaxy (MBE). To achieve this, we apply single-pulse four-beam laser interference during the epitaxial growth step. The interference pattern generates transient photothermal gradients at the...
nanoscale on the growth surface, resulting in the formation of spatially organized nanoislands. We suggest these are formed by adatom diffusion towards the colder regions of the pattern at the interference minima. The subsequent deposition of InAs on such a structured surface leads to the controlled nucleation of QDs, in which the edges of nanoislands create energetically preferential nucleation sites. By adopting optimized laser parameters and appropriate growth conditions, e.g., laser polarization, InAs coverage and growth rate of InAs, dense ordered arrays of single QDs or QD molecules with a pitch of 300 nm can be obtained. The patterned QD ensembles show narrow and bright photoluminescence (PL), indicating good optical quality and uniformity. Here, the detailed experimental processes including the growth protocol and laser interference patterning methodology will be discussed.

EXPERIMENTAL DETAILS

The growth experiments were carried out with a conventional MBE system combined with in-situ single-pulse laser interference patterning. The laser interference system uses a flash-lamp pumped Nd:YAG laser source (Innolas Spotlight) operating on the 3rd harmonic at an output wavelength of 355 nm, a pulse duration of 7 ns, a Gaussian beam diameter of 5 mm, a repetition rate of 5 Hz and with pulse energy in the range of 10-40 mJ. After beam splitting, four coherent beams were guided upward into the MBE chamber at an incidence angle of 58° via four symmetric anti-reflective optical vacuum viewports. The beams then recombine at the center of the substrate. The ultraviolet (UV) beams are viewed from the luminescence of an InGaN/GaN wafer on the sample holder, which allows us to observe a blue-green image of the spots with an upward facing camera. Since the beams are projected with a large angle of incidence, it is observed that the overlapping area of the four laser beams is reduced to around 3 mm in diameter. The interference pattern from this configuration can be calculated and consists of a 300 nm square grid of high and low laser fluence spots. The nature of the interference pattern has been confirmed by ex-situ lithography studies on photoresist using an equivalent geometry.

The InAs QD samples were fabricated on 2-inch epi-ready n-type GaAs(100) wafers. After oxide desorption, a 500 nm GaAs buffer layer was grown at a substrate temperature of 600°C, after which the substrate was cooled to 500°C. One monolayer (ML) of InAs was then deposited at different growth rates (0.079 ML s⁻¹, 0.04 ML s⁻¹ and 0.026 ML s⁻¹), immediately followed by in-situ single-pulse laser interference patterning. Then, a further deposition of InAs ranging from 0.5 ML to 0.75 ML was supplied. The initial 1 ML of InAs is below the critical thickness and the additional deposition takes the InAs coverage up to and beyond the critical thickness for the 2D to 3D transition. We have observed qualitatively similar results if we apply the laser pulse earlier or later in the deposition cycle. During the laser patterning, the substrate rotation was stopped. After 10 seconds of growth interruption under As₂ flux, the samples were quenched and taken out from the chamber for structural characterization.

RESULTS AND DISCUSSION

The surface morphologies were investigated ex-situ by atomic force microscopy (AFM) using an FSM NanoView-1000 instrument. Figures 1(a-f) present AFM micrographs of a sample with a total of 1.55 ML InAs coverage grown on GaAs at a growth rate of 0.079 ML s⁻¹. The laser pulse energy used for patterning is 40 mJ and the polarized angle of the four beams is 58°. The evolution of the pattern is associated with a laser intensity variation which is a primarily a consequence of the overlap of Gaussian beams. There is also a weak Moiré effect, generated by small angular offsets.
A schematic diagram of the four-beam laser interference configuration is shown in Figure 2(a). Square arrays of symmetric nanoislands with a pattern pitch of 300 nm are clearly observed in Figures 1(a-d) and the cross-sectional profiles along the [100] and [010] directions are shown in Figure 1 (g-k). The formation of these nanoislands originates from interference-induced surface diffusion at the nanoscale according to our simulation results. Figure 2(b) schematically illustrates the formation processes of nanoislands and QDs, where firstly the laser interference is applied after 1 ML of InAs deposition. Owing to the absorption of the incident UV pulse, heat is generated at the near surface of the wafer. The temperature rises rapidly (~ns) at the interference maxima producing a strong lateral thermal gradient which acts as a driving force for atoms to migrate from hot regions (interference maxima) towards adjacent cold regions (interference minima). Illustrated in the direction of arrows, where islands then form. By continuously depositing a further 0.55 ML of InAs, QDs can be nucleated at the island sites, which is clearly observed in Figure 1(d).

Figure 1(a) shows the simulated interference pattern depicted in the inset. Large square-shaped islands with star-shaped extensions are formed, with a shallow pit of 0.7 ± 0.1 nm in depth in the center, which may be ascribed to an insufficient diffusion length across the broad islands under effects such as strain. Away from the center of the beam overlap, where the pulse energy is lower due to the Gaussian distribution of the laser beams, we observe a smaller island size. This can be observed in the trend from Figure 1(b-d). Square-shaped islands with a reduced central pit depth of 0.4 ± 0.1 nm are shown in Figure 1(b). In regions of lower pulse energy, the islands are smaller and are without a central pit; typically 100-150 nm in width and with a height remarkably consistent at 1.0 ± 0.2 nm (2-3 ML), as shown in Figure 1(c). In Figure 1(d) we have the smallest stable island sizes we have observed of 70 nm width. We attribute this reduction in width to a lowering of the pulse energy. Away from the center of the beam overlap we have a lower energy and a smaller thermal effect. In these conditions fewer surface atoms can diffuse and therefore the smaller the islands become. Further local changes in interference pattern and beam energy occur as a consequence of a Moiré effect.

The results suggest that above a certain critical temperature, which is a combination of the growth temperature and the temperature rise due to the absorbed laser pulse, sufficient thermal energy is gained to overcome the diffusion barrier for adatom migration. At present, the composition of the nanoislands cannot be confirmed due to the difficulty of obtaining compositional analysis on exceedingly shallow and small (<100 nm) features. However, we speculate that the nanoisland is mainly composed of indium atoms, since these are relatively mobile compared with gallium atoms and will have a lower diffusion barrier and therefore a lower energy threshold.

The profiles of nanoislands along the [100] and [010] directions in Figure 1 (g-k) show small dips of 0.5 ± 0.1 nm at the boundary around islands, which appears as a dark ring in the AFM images. The origin of the surface dip may be related to the local removal of material by enhanced diffusion of adatoms owing to the strain field around the islands, or to the local decomposition since surface adatoms at kinks on the steps are more loosely bound than terrace atoms. The negative surface curvature present in this situation is expected to provide energetically favorable positions for the nucleation of QDs, since adatom diffusion is always driven by the gradient of chemical potential. The nucleation of QDs at downward steps on pre-patterned substrates using nano-holes or trenches has been previously reported. For the growth on such a nanoisland...
structured surface, it is expected that the nucleation of QDs will be mainly determined by the surface morphology. This is indeed what we observe and nucleation of QDs at the edge of the nanoislands has been found reliably in more than 30 samples. As shown in Figures 1(d) and (e), the probability of finding a QD is inversely proportional to the size of the nanoislands. With decreased island size, more indium atoms can accumulate within the very concentrated area, giving rise to a high nucleation rate for QDs as the coverage of InAs reaches the critical value (~1.6 ML).21 Large QDs of a more consistent size can then be formed due to the coalescence of small dots or through Ostwald ripening.28

Typically, large QDs with a width of 40-50 nm and height of 12-15 nm are observed, as demonstrated in Figure 1(e). It is also found that as the QDs form, the nanoislands gradually disappear and it is therefore assumed that the growth of large QDs is at the expense of material coming from the nanoislands. The variation of QD occupancy at each site is attributed to nanoisland size fluctuation which may be a consequence of an unintentional laser intensity variation. In the area between adjacent islands, the nucleation of interstitial QDs was found to be completely suppressed owing to the enhanced surface diffusion of deposited InAs. Compared with the non-patterned region (Figure 1(f)), in which small QDs are randomly nucleated and have a bimodal size distribution with a density of $1 \times 10^{10}$ cm$^{-2}$, indicating that the InAs coverage is below the optimal value for a homogeneous distribution, the patterned region exhibits a much lower QD density (~2×10$^9$ cm$^{-2}$) with enlarged dots and better size uniformity. The results suggest that indium diffusion is strongly promoted to reach the critical thickness at the interference minima whilst being largely suppressed on the planar areas between the pattern sites. We note that indium desorption under the thermal influence of the laser interference has been suggested previously to be the origin of the QD patterning process. This is not consistent with the larger QD size and density we observe in the patterned regions which come from a local enhancement in the indium density at the interference pattern minima and not from desorption at the pattern maxima.

With different InAs coverage, we observed a noticeable influence of the deposition amount on the number of dots per island site. Figure 3 displays three-dimensional AFM images of sample A-D with the same growth conditions and laser parameters, but with different InAs coverages: 1.55 ML, 1.60 ML, 1.65 ML and 1.75 ML, respectively. At subcritical InAs coverage, single or pair QD occupancy is dominant. By increasing the amount of InAs deposition, the average number of dots grown at each site changes from 1 to 6 and can be more with additional deposition. For these larger amounts of deposited InAs QDs can nucleate close to each other to form QD molecules which are almost symmetrically arranged around the nanoislands. The potential electronic coupling between their wavefunctions makes these structures promising as entangled photon sources for quantum cryptography and quantum computation.6,7,29-31 It should also be noted that when we increase the InAs coverage to 1.8 ML, the number of QDs per site increases and additionally we observe the coalescence of adjacent QDs and the formation of a few dots on the planar areas between the pattern sites. The AFM image of this situation is presented in the Supporting Information. Hence, the deposition amount of InAs is critical for the control of QD number per site. We can see that a slight change in InAs coverage has a dramatic effect on the dot occupancy and that controlled low amount of InAs supply is capable of achieving arrays of single QDs.

The laser interference parameters also have a strong impact on the formation of nanoislands and QDs. The interference pattern varies with laser parameters such as angle of incidence, azimuth angle and polarization state.20 We have changed the polarization angle of the laser beams to investigate its effects. Figure 4 compares the surface morphology of two samples grown under same conditions, but with two different polarization states; one with a polarization angle of 58° and the other with 90°, obtained by a rotation of the polarization using a half-wave plate and polarizer in each beam path. For both samples, the growth rate has been lowered to 0.04 ML s$^{-1}$ in an attempt to reduce the QD density. From the AFM image in Figure 4(a), the nanoislands formed at a polarization angle of 90° are relatively larger in width and lower in height than those at 58°. This can be explained by the simulation results shown in the insets. The cold regions of the photothermal pattern in Figure 4(e) are more closely confined by hot regions than those in (a) and therefore the situation with a polarization angle of 58° is more conducive to the formation of concentrated small nanoislands. Moreover, no surface dip around islands was observed in Figure 4(a) since the
laser pulse energy in this (essentially all TE) polarization state is much lower due to strong reflection from the GaAs surface. Hence, insufficient thermal energy results in relatively few diffusing In atoms at the interference maxima and the strain between the islands and the layer surface also reduces. Consequently, the lack of good nucleation sites in the 90° polarization case results in relatively higher density (~5 × 10^9 cm^-2) and a very dispersed distribution of QDs with size fluctuation ranging from 4-16 nm in height (Figure 4(b) and (d)). In contrast, highly ordered QD arrays of single dot or pair occupancy with a lower density of 1×10^9 cm^-2 are realized at the polarization angle of 58°, as presented in Figure 4(f). The corresponding dot occupancy statistics of two samples are presented in Figure 4(c) and (g), and QD height statistic distribution displayed in Figure 4(d) and (h), respectively. The heights of QDs shown in Figure 4(h) range from 10-16 nm with the distribution centered at 13 nm. The precise control of the interference parameters, particularly the polarization, has a significant influence on the QD arrangement, especially its effect on the formation of the surface dip, which is shown to be critical for the controlling of QD nucleation. To further optimize the patterning process, the effects of other experimental conditions were investigated.

To obtain better QD size homogeneity, a lower InAs growth rate of 0.026 ML s^-1 was employed to enhance the adatom migration length. Meanwhile, we reduced the laser pulse energy to 10 mJ and

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Figure 5. 3-D AFM micrographs of (a) sample A, (b) sample B and (c) sample C with 1.5 ML InAs on GaAs(100) substrates. (d) AFM image of non-patterned region of sample A. (e) QD height distribution of sample A. The inset is the corresponding 2D AFM image of QD arrays.

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Figure 6. Excitation power-dependent PL spectra of an InAs QD array at 88 K. The inset shows the logarithm plot of I_{PL} as a function of P_d. The slope k=0.95.
compared the results of applying the pulse earlier or after the deposition cycle. Figure 5 shows AFM micrographs of three samples with the identical deposition amount of 1.5 ML InAs on GaAs(100) substrates but with different exposure-growth sequences, in which Figure 5(a) represents the sample A of applying the laser pulse after depositing 0.5 ML InAs, immediately followed by additional 1 ML InAs deposition; Figure 5(b) shows the sample B of the laser pulse introduced between the deposition of 1 ML and 0.5 ML InAs; and Figure 5(c) demonstrates the sample C where the pulse was applied after the total 1.5 ML deposition of InAs. It is clear that all these three samples are capable of fabricating single dot arrays, albeit with different QD size fluctuations. Sample A has the best size homogeneity, while sample C shows a large size variation. It appears that after the laser patterning, more InAs is needed for sufficient uniform migration to achieve uniform growth of individual QDs. The QD height statistical distribution of sample A is illustrated in Figure 5(e), with an average diameter and height of ~70 nm and ~14 nm respectively. The relatively large size of QDs than that in Figure 4 is due to the slower growth rate. For comparison, Figure 5(d) displays the non-patterned region (edge of sample A), which contains only shallow quasi-3D islands, indicating that the InAs coverage is below the critical thickness. It is also noticeable that in sample C we can still see QDs at such a low InAs coverage, rather than the quasi-3D islands formed in the non-patterned region. Moreover, we observed that the nanoisland structures in sample C and the InAs 2D monolayers still remaining on the planar regions between those island sites (see the Supporting Information for details). All these observations prove that the nanoisland and QD formation is driven by diffusion instead of desorption. Laser interference patterning induces the nucleation and growth of QDs below the normal (unstructured) surface critical thickness value for the transition and improves the size homogeneity. In addition, by using lower pulse energy, we can extend the formation of the smaller nanoislands to a larger surface area. The area for which we can presently achieve highly ordered single QD arrays is around 1 mm². Further expansion is limited at present by the Gaussian beam profiles. However, this is not a fundamental limitation and with suitable laser optics could be addressed.

The optical properties of the patterned QDs are investigated by low-temperature ensemble-photoluminescence (PL) spectroscopy. Samples for PL characterization were capped with a 200 nm thick GaAs layer and with an additional 300 nm layer of AlGaAs grown prior to GaAs buffer in order to enhance the PL signal. The samples were mounted in a continuous flow cryostat cooled to 88 K with liquid nitrogen. A 659 nm continuous-wave pump laser was focused through a 20× objective to around 10 µm spot on the sample. The PL signal was spectrally resolved by a spectrometer and a liquid nitrogen-cooled InGaAs detector. Figure 6 shows the excitation power-dependent PL spectra of sample A at 88 K. Strong PL signals at the ground state (GS), the first excited state (1ES) and the second excited state (2ES) emissions are clearly observed, where the peaks are at 1.05 eV, 1.12 eV and 1.17 eV, respectively. The inset of Figure 6 presents the logarithm plot of integrated PL (IP) intensity of the ground state emission as a function of excitation power density (P). The fitting slope (k=0.95) is close to unity (~1) at 88 K, according to the power law, the integrated PL intensity increases with the excitation power density, indicating that radiative recombination dominates the recombination process. These results are comparable to those reported results of high-quality self-assembled InAs QDs with a low growth rate. A very narrow linewidth of 22 meV for the GS peak is observed at low power, which is smaller than most values previously reported by using other nanopatterning technologies. It appears that the size variation of these dots are reasonable small, indicating their good crystalline quality. This suggests that the laser interference does not degrade the optical quality of the QDs and this observation may enable the implementation of optically efficient quantum devices using this scheme.

CONCLUSION

We have demonstrated that high structural and optical quality QD arrays can be grown by a combination of in-situ laser interference patterning and self-assembled epitaxial growth. Distinct regimes of nanoisland and QD formation were observed during the growth of InAs/GaAs(100) by MBE. Our results show that single pulse interference on the growing surface is capable of manipulating the atomic kinetics to promote QD nucleation. Periodic arrays of nanoislands yield energetically favorable nucleation sites for QDs which nucleate at the edge of the islands. Excellent lateral ordering of single QDs or QD molecules has been obtained, guided by the presence of the nanoislands. Although our results are specific to III-V MBE, the technique can be applicable to other materials and growth technologies which have thermally activated growth processes. This novel technique provides a fast and efficient means for providing highly ordered arrays of nanostructures and offers a template for the realization of single QDs or QD molecules for future quantum information technology device applications.

ASSOCIATED CONTENT

Supporting Information

AFM micrograph of the sample surface with 1.8 ML deposition of InAs on GaAs(100) substrate. AFM micrograph of the sample C surface with 1.5 ML deposition of InAs on GaAs(100) substrate followed by laser interference patterning.

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All authors have made equal contribution to the work.

Notes

The authors declare no competing financial interests.

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