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14. ABSTRACT
One of the biggest stumbling blocks in semiconductor nanotechnology today is the inability to manufacture quantum dot (QD) arrays with uniformity of QD size and spacing. This report describes a comprehensive research program to achieve dense (?1E10 cm^-2) QDs with uniform size and spacing, over large areas, with luminescence suitable for optoelectronics device applications. The descibed techniques require only easily available equipment, and are inherently parallel and suitable for high-volume production. The method to achieve regularly-spaced QD

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Report Title

Hyper-uniform site-controlled quantum dot arrays prepared by soft-imprint lithography technology

ABSTRACT

One of the biggest stumbling blocks in semiconductor nanotechnology today is the inability to manufacture quantum dot (QD) arrays with uniformity of QD size and spacing. This report describes a comprehensive research program to achieve dense (?1E10 cm^-2) QDs with uniform size and spacing, over large areas, with luminescence suitable for optoelectronics device applications. The descibed techniques require only easily available equipment, and are inherently parallel and suitable for high-volume production. The method to achieve regularly-spaced QD arrays relies on soft nanoim-print lithography to pre-pattern a semiconductor (GaAs) substrate into nanometer scale pores array with sub-100 nm periodicity, followed by regrowth of (InAs) QDs on top. The uniform nano-pore pattern on GaAs causes InAs QD nucleation only at designed loca-tions with uniform size, leading to an array of uniform-sized site-controlled QDs (SCQD). The proposal addresses specific challenges that preliminary study of 200nm pitch SCQD arrays has identified, along with studying and managing the complex interplay of all the process variables to get an optimal result.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

1. Chien-Chia Cheng, K. Meneou, and K. Y. Cheng, "High optical quality InAs site-controlled quantum dots grown on soft photocurable nanoimprint lithography patterned GaAs substrates," Applied Physics Letters, v.95, p.173108 (2009)
2. Chien-Chia Cheng, K. Meneou, and K. Y. Cheng, "Molecular-beam epitaxy growth of site-controlled InAs/GaAs quantum dots defined by soft photocurable nanoimprint lithography," J. Vacuum Science and Technology B, v.28, p.C3C37 (2010)

Number of Papers published in peer-reviewed journals: 2.00

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Patents Submitted

Patents Awarded

Graduate Students

NAME	PERCENT_SUPPORTED
Chien-Chia Cheng	0.25
Kevin Maneou	0.25
FTE Equivalent:	**0.50**
Total Number:	**2**

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

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The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME
Kevin Meneou

Total Number: **1**

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:

Total Number:

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Final Report

Hyper-uniform site-controlled quantum dot arrays prepared by soft nano-imprint lithography technology

By

Prof. K. Y. Norman Cheng
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The main focus of this research is to develop a manufacturable soft nano-imprint lithography method to fabricate site-controlled quantum dots (SCQDs) with high dot density, uniform size distribution, and excellent optical quality such that quantum dot (QD)-based photonic devices with performance approaching theoretical predictions can be realized. The detailed tasks and results are listed below.

1. 200 nm pitch soft lithography technology development:
   - Replicate 200 nm pitch/l00 nm hole patterns onto GaAs substrate
   - Optimize pattern transfer for fidelity and uniformity of feature dimensions

Over the past two decades, there has been great interest in integrating self-assembled quantum dots (SAQDs) into electronic and optoelectronic devices, utilizing three-dimensional quantum confinement effect to improve device performance as well as create devices with added functionalities. However, SAQD growth via the Stranski-Krastanov (SK) growth mode does not provide much control over QD size, spatial position, and uniformity. Consequently, improving homogeneity and spatial distribution of QDs become the next important issues in QD research, which lead to the concept of site-controlled quantum dots (SCQDs). Since the late 1990s, SCQDs have been realized through the growth on patterned substrates prepared by electron beam lithography (EBL) and focused ion beam (FIB). Although improved QDs size distribution and narrowed photoluminescence (PL) linewidth at very low temperatures have been accomplished, devices with improved optical performance have not been demonstrated. Furthermore, due to the serial nature of EBL and FIB lithography, the cost would be extensive if large amount of devices need to be fabricated. As a result, it is necessary to develop alternative approaches for the fabrication of high optical quality SCQDs cost efficiently. Using self-organized pores in nano-channel alumina to define nucleation sites of the SCQDs, room temperature PL has been demonstrated. However, challenges remain in improving the ordering of the nanopores on the profiled GaAs substrates and the percentage of QDs that stack inside the nanopores.

In this program, we developed a pathway to overcome problems mentioned above by utilizing soft photocurable nanoimprint lithography (soft NIL) to create patterns of uniform nanoscale sites for QDs growth. NIL is promising for high-throughput and low-cost nanostructure fabrication through parallel patterning. Most importantly, the soft NIL pattern transfer process used in this study is achieved by deforming the photoresist (PR) through physical contacting, no high-energy beam is involved, and thus, avoiding potential damage to the substrate.

The (100) semi-insulating GaAs substrate is coated with 50 nm silicon dioxide followed by a UV-curable PR of 120 nm thick. Next, the soft NIL technique is used to replicate the designed pattern in the silicon dioxide and PR layers. The pattern used in this study is a large area (several cm^2 square) array of 100 nm × 100 nm square nanopores on a 200 nm pitch, yielding an array density of 2.5E9 cm^-2. The residual PR within each nanopore and underlying silicon dioxide are removed by reactive ion etching (RIE) using oxygen and Freon gases, respectively, at 150 W radio frequency (RF) power. Then the pattern is transferred to GaAs substrate by wet etching. The wet etching step is especially important for removing residual ion-related damages within nanopores. A solution of ammonium, hydrogen peroxide, and deionized water mixture is used for wet etching. Finally, silicon dioxide with residual PR is stripped by hydrofluoric acid under ultrasonic agitation. In order to achieve high QD quality, a set sequence of cleaning steps is carried out on all patterned samples before molecular beam epitaxy (MBE) growth. To begin with, samples are treated by oxygen plasma, followed by ultrasonic cleaning in acetone, methanol, and isopropanol. Then, a sequence of hydrochloric acid and sulfuric acid etching is applied, intended to remove surface oxides and passivate the surface, respectively. Finally, surface treatment procedure ends with a thorough deionized water rinse and nitrogen blow dry. Afterward, samples are loaded in a MBE system for QD growth. Atomic hydrogen-assisted desorption of surface oxides is used inside the growth chamber prior to MBE growth. Thermally cracked atomic hydrogen is applied for an hour with the samples held at 480 °C. After desorption, all samples are covered with a 30-monolayer (ML) GaAs buffer layer grown at 500 °C, followed by 3 ML of InAs with a growth rate of 0.014 MLs deposited at the same temperature. After the deposition of InAs QD layer, a 120 s growth interruption under a constant arsenic flux is inserted to enhance the formation of QDs. Finally, the sample is cooled to room temperature for surface morphology characterization and PL measurements. The nanopore array patterns are transferred using wet etching in parallel fashion from the silicon master into GaAs with high fidelity. In addition, very low defect density such as missing nanopores is observed over a large area scanned. Atomic force microscope (AFM) surface image determines the etching depth of nanopores is 15 nm while maintaining the original square shape with smooth edges.

2. MBE regrowth of SCQDs:

- Determine growth window where SCQD growth occurs
- Optimize for luminous intensity, uniform QD size.

After soft NIL pattern transfer and ex situ cleaning steps, the GaAs sample is loaded into the MBE system for QD growth. As the dimension of patterns shrinks down to nanoscale, extra care must be paid during the in situ surface clean process to retain pattern integrity. It has been shown that the conventional thermal desorption used to eliminate surface contaminants can severely distort the nanoscale patterns even if the duration of high temperature treatment is short. Therefore, a low temperature atomic hydrogen-assisted cleaning process is adopted in this study. For nanopatterned sample surface after atomic hydrogen-assisted cleaning, AFM scans indicates the depth profiles are almost unchanged from the wet etching prepared sample. This fact confirms the ability of preserving nanoscale patterns by the low temperature hydrogen-assisted desorption approach. The edges of nanopores become slightly blunt after atomic hydrogen-assisted cleaning, but it does not have a noticeable influence on SCQDs formation. After in situ oxide desorption, a 30 ML GaAs buffer layer followed by 3 ML of InAs is deposited. No QDs are formed on the flat surface between nanopores, suggesting the growth parameters for SCQDs formation are appropriate. Furthermore, one single dot inside each nanopore is demonstrated on the wet-etched sample, and the average diameter and height is 63 and 9 nm, respectively. Next the PL spectra from wet-etched SCQD sample as well as an unpattemed SAQD reference sample are compared. The

SAQD reference sample is prepared by mounting a piece of unpatterned epi-ready GaAs onto the MBE sample holder alongside the wet-etched SCQD sample. The two samples undergo oxide desorption and MBE growth together, resulting in SAQDs on the unpatterned piece of GaAs and SCQDs on the patterned piece. The PL peak position of the SCQD sample is 0.932 eV, compared to 0.792 eV for the SAQD case. The shorter peak wavelength for SCQDs originates from the fact that SCQDs has a wider base and a shorter height than SAQDs. The average height of SCQDs is 6 nm, compared with that of 8 nm in SAQDs. Moreover, the full-width-at-half-maximum of the PL peak of the SCQD sample is 102 meV, which is 11 % narrower than that of SAQD sample and attributed to the more uniform QDs on patterned substrates. The integrated PL intensity from the SCQD sample is as strong as that of the reference SAQD sample by taking the dot density into consideration where the dot density on SCQD sample (25 /µm^2) is about half of that on SAQD samples. The ratio of integrated PL intensity between 300 and 77 K remains the same on both SCQD and SAQD samples. This confirms that the optical quality of regrown SCQDs layer on processed samples is comparable to that of SAQDs grown on unprocessed epi-ready substrates.

In conclusion, InAs site-controlled QDs have been demonstrated on (100) GaAs substrate patterned by soft photocurable NIL and regrown by MBE. By judicious use of wet etching and atomic hydrogen-assisted desorption instead of more conventional techniques, optically active, uniform QD arrays are achieved with a single QD at each designed nucleation site. Room temperature PL of SCQDs from wet-etched sample shows an integrated intensity comparable to that of SAQDs, indicating a defect-free etched interface and a high quality QD growth. A narrower PL linewidth from the SCQD sample is also an indication of a more uniform QDs distribution on soft NIL patterned substrates. These features suggest that using soft NIL technique to transfer nanoscale patterns is an excellent approach of achieving highly uniform QDs with precise positioning and good optical quality at a low cost and high throughput for future device applications.