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Deterministically Placed Quantum Dots for Quantum Nanophotonics

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ABSTRACT

The ability to achieve deterministic placement of semiconductor quantum dots (QDs) opens up interesting possibilities for nanophotonic devices. By incorporating these QDs within microcavities, light-matter interaction can be tailored and enhanced, enabling phenomenon such as spontaneous emission enhancement, low threshold lasing, single photon emission and strong-coupling. The quality of these phenomena relies on the distribution of emission wavelengths of the emitter dipoles and the strength of their coupling to internal fields of the cavity. Therefore size-controlled fabrication of QDs and their deterministic placement become quite important. In this work we will describe a photoelectrochemical-based etching of III-nitride materials to achieve QDs with uniform emission wavelength. By patterning using electron beam lithography to create a nanopost structure in an epitaxially grown III-nitride based quantum well structure, we will show potential for deterministic placement. The photoluminescence response from the nanopost structure after photoelectrochemical etching reveals sharp lines indicative of quantum dot formation.

Keywords: Quantum dots, photoelectrochemical etching, photoluminescence.

1. INTRODUCTION

Nanostructures have become ubiquitous in modern nanophotonics and nanoelectronics. In particular, semiconductor quantum dots (QDs) have become important as nanoscale light sources in photonic applications [1-5]. QDs are less than ~10 nm which is on the order of exciton Bohr radius for many semiconductors where quantum-size effects become prominent. For QDs the optical absorption/emission wavelength depends on the size of the QD. Furthermore, the quantum size effects results in discrete "atomic-like" energy levels turning QDs into nanoscale atoms with size tuneable emission wavelength. Several different approaches exist currently to fabricate QDs including those that are solution based. Nevertheless, a nanofabrication-based approach, either bottom-up epitaxial self-assembly (e.g. Stranksi-Krastanov (S-K)) or top-down etching are likely to be more suitable for semiconductor-based integrated photonics applications. However, two main challenges remain, namely achieving uniform size distribution for ensembles of QDs so as to achieve uniform emission wavelength and deterministic placement of QDs in nanostructures. The S-K based approach typically yields a large size distribution accompanied by uncertainties in the location of such quantum dots. This makes yields of nanophotonic devices quite low as many devices either may not have the required quantum dot size or the required placement. One approach to achieve a uniform emission wavelength from QDs has been recently demonstrated [6] on epitaxial structures. This is achieved by quantum-size controlled photoelectrochemical (QSC-PEC) etching which is similar to the technique used in earlier works [7, 8] on colloidal solution-based QDs. The approach utilizes the enhanced wet etching in acid due to light-assisted carrier generation in the semiconductor that self-terminates when quantum size effects set in and suppress light absorption. The previous approach has demonstrated this in an unpatterned epitaxial GaN film containing a single InGaN quantum well which resulted in QDs with random placement. In this work we will utilize QSC-PEC etching to create QDs with deterministic placement by pre-patterning the epitaxial film using electron beam lithography. We will present optical spectroscopy from PEC-etched III-nitride nanoposts. The ability to achieve deterministic placement of QDs opens up interesting possibilities for nanophotonic devices such as single photon sources by coupling to high quality factor microcavities which can be formed more precisely around a known QD location.

2. FABRICATION OF QUATUM DOT IN NANOPOST

Three-nitride materials are difficult to wet etch using acid or base solutions. Enhanced carrier generation through photo excitation can substantially improve wet etching in III-nitrides. A typical set-up for photoelectrochemical (PEC) etching is shown in Figure 1a. The etch process is bandgap selective, dopant selective and light intensity dependent and can thus be controlled and monitored via the etch current flowing between the electron and the material[6]. When a relatively narrow linewidth (~ 1-2 nm) source such as filtered lamp source (e.g. Xe arc lamp) or a laser source (tunable ps Ti:sapphire) with wavelength shorter than that of the semiconductor band gap

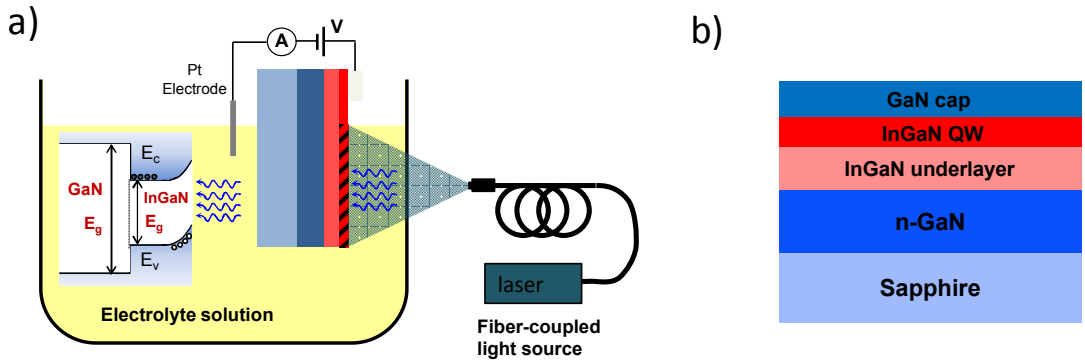

Figure 1. a) Schematic of the photoelectrochemical etch cell. b)Planar epitaxial III-nitride structure used for PEC etching.

is illuminated on the sample, electron-hole pairs are generated. The PEC etch process begins with semiconductor surface oxidation of photoexcited holes[9, 10] which is etched away by the acid solution. As the size of the QD continues to reduce during the etching process, quantum size effects emerge causing the electronic bandgap to increase. Beyond a certain critical QD size, the bandgap becomes larger and the input light is no longer absorbed and the etch thereby terminates. A typical metal-organic vapor phase epitaxy (MOVPE) grown epitaxial structure used for this work is shown in figure 1b. The structure consists of ~ 4 µm thick GaN layer grown on a sapphire template. This is followed by 185nm of $In_{0.04}$ $Ga_{0.96}$ N underlayer followed by 2.7nm thick $In_{0.15}$ $Ga_{0.85}N$ quantum well layer followed by 10 nm of GaN cap layer. When the starting substrate is unpatterned the etchant enters through micro/nanoscale defects in the thin GaN cap layer and etching proceeds laterally resulting in non-uniform distribution of dots.

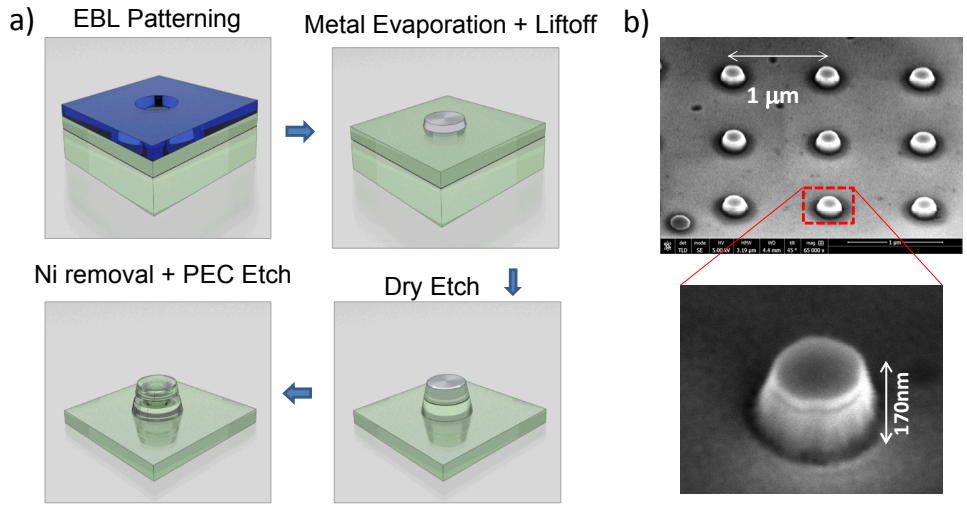

Figure 2. a) Schematic of nanopost fabrication. b)Scanning electron microscopy(SEM) image of the nanopost structure.

For this work we show that we can fabricate QDs at specific locations in a nanostructure. We start out by first creating a patterned template to create a nanopost. The process schematic is shown in figure 2. We first create a pattern on a polymethylmethacrylate (PMMA) resist spun on the epitaxial structure shown in figure 1b using electron beam lithography. The pattern consists of an array of 100-200nm diameter circles spaced 1 µm apart which leaves holes or openings upon resist development. The openings are then filled with 20-30nm of Ni deposited via electron beam evaporation followed by lift-off. This leaves behind nickel "dots" that act as a hard mask. The lift-off is followed by an inductively coupled plasma (ICP) etch utilizing a Cl_2/BCl_3-based etch chemistry. Addition of an initial step where a set of alignment marks is patterned, enables subsequent cavity structures (e.g. photonic crystal cavities) to be fabricated in precise alignment to the QDs in the nanopost. The dry etch is followed by a PEC etch to form QDs in the InGaN quantum well layer. The PEC etch was performed for ~ 90 minutes using the etch cell shown schematically in figure 1a in a 0.2M H2SO4 solution at an illumination wavelength of 430nm where the source was fiber-coupled into the cell from a laser source. The

laser source is a frequency doubled Ti: Sapphire pulsed laser with a ~ 2ps pulse at an 82 MHz repetition rate with an average power of 30mW. Figure 2b shows a scanning electron microscope image of a PEC-etched nanopost. The magnified image shows a dark ring just below the surface where the solution has preferentially etched into the InGaN quantum well.

3. OPTICAL SPECTROSCOPY ON THE QD IN NANOPOST

We performed low temperature photoluminescence (PL) measurements from the PEC-etched posts to obtain spectroscopic information on the QD formation. The sample was housed in a cryostat nominally cooled to 10K. The sample was pumped at a wavelength of 375nm using a frequency-doubled Ti:Sapphire pulsed laser with ~ 2ps pulse width at an 82MHz repetition rate. The laser with an average power of 5 μW was focused to a spot nominally about 1μm in diameter using a microscope objective. The objective also served to collect the PL from the QD in the nanopost. Due to vibration of the cryostat during operation, combined with the small spot size, PL measurement required shutting the cryostat off for a short period while measuring PL to minimize any smearing of the emission features. This resulted in slight increase in temperature (~ 10 K) during the measurement window, but allowed us to record spectra showing evidence for QDs Figure 3 shows a PL spectrum from a PEC-etched post with a nominal diameter of 150 nm. We observe three small sharp peaks at 409 nm, 411.6 nm and 414 nm on top of a broader PL emission between 406 and 420 nm. The narrow linewidths of the peaks suggests the formation QDs inside the nanoposts, however, the three distinct peaks also suggests formation of multiple QDs. Possible reasons for this include relatively large diameter (150 nm) compared to the thickness of the capping GaN layer (~ 10nm) which might result in etching from the top surface in addition to the sides. It is known that etching can be done through a 10nm thick GaN cap layer[6]. The PEC-etch through the exposed sidewall of the post, which is a faster etch and the desired, competes with etching of nitride material that takes place through the GaN cap layer. These competing PEC etches likely results in multiple dots in a single post. In

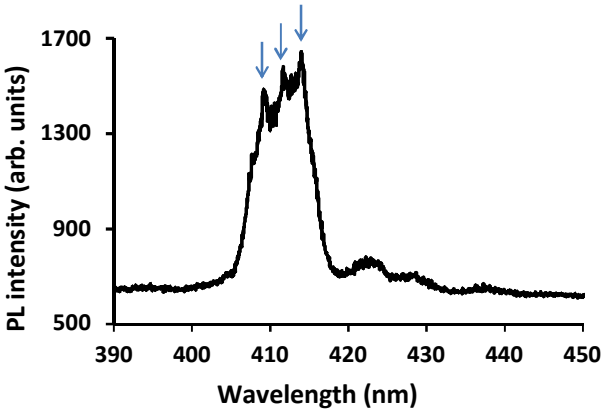

Figure 3. Photoluminescence spectrum from a PEC etched nanopost with 150nm diamter. Three peaks indicating possible quantum dot formation.

the future, we plan to use posts made from a thicker GaN capping layer (> 30nm) for PEC etching as well utilizing a cryostat with dramatically reduced vibration.

4. CONCLUSIONS

We have shown a photoelectrochemical etch based approach to achieving quantum size-controlled quantum dot formation starting with an epitaxially grown III-nitride film. By using electron beam lithography patterning, nanoposts can be created in a deterministic fashion that can be PEC-etched to create QDs in order to subsequently enable creation of microcavities to control QD emission. The PEC etched QDs exhibit sharp lines in their photoluminescence spectrum indicating formation of QDs. Fabrication of QDs with controlled emission and deterministic placement will be enable many interesting future quantum nanophotonic devices.

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