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Ultra-low surface recombination for deeply etched III-V semiconductor nano-cavity lasers

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Abstract: We investigated the passivation of III-V semiconductor nanostructures using wet-chemical ammonium sulfide treatment and SiO_x encapsulation. We achieved an ultra-low surface recombination velocity value of ~530 cm/s enabling the future development of high-performance room-temperature nanolasers.

OCIS codes: (140.0140) Lasers and laser optics; (160.0160) Materials; (250.5300) Photonic Integrated Circuits; (250.5980) Semiconductor Optical amplifiers

1. Introduction

In the last few years, many nanoscale light sources have been reported, such as nanocavity lasers employing III-V materials as the gain medium [1, 2]. Scaling down the laser sources to the size below their operation wavelength (< 1.5 μm) will greatly contribute for the deployment of on-chip optical interconnects thanks to their small footprint, low energy consumption and high speed [2]. Despite recent efforts, difficulties in achieving room temperature (RT) and continuous wavelength (CW) operation with high output optical power considerably limits the use of nanolasers in future optical communication systems. Among many challenges, non-radiative processes including surface recombination have been shown to have a detrimental effect in the efficiency of deeply etched *pn*-junctions such as in sub-wavelength cavity nanolasers [2]. Due to the high surface-to-volume ratio of such nanostructures, large areas of the active material are exposed with a high density of non-radiative recombination centers formed from the bombardment of high energy ions during etching. As the size of the active region becomes comparable with the carrier diffusion length, surface effects begin to strongly influence their performance. Since the non-radiative processes reduce the radiative efficiency, the threshold current of a nanolaser increases as the injected carriers recombine faster [3]. Therefore, the reduction of non-radiative centers is a key factor for enabling RT at CW operation in nanolasers.

Although surface recombination strongly limits the performance of sub-μm devices, passivation treatments of the surface of nanoscale III-V semiconductor heterostructures have remained almost unexplored apart from a recently published photonic crystal nanolaser [1,3] reporting a 3-fold reduction of the surface velocity ($S=4 \times 10^3$ cm/s) when compared to best values obtained in [4] ($S=1.2 \times 10^4$ cm/s). Here, we report ultra-low surface velocities for sub-μm III-V deeply etched nanopillars using a sulfide passivation treatment that comprises a gentle wet etching of the active material plus ammonium sulfide solution at 20% followed by a thin SiO_x capping. We observed a very robust passivation effect that enhances the photoluminescence (PL) up to two orders of magnitude compare with un-passivated samples. Time-resolved microPL measurements further confirm the substantial improvements showing two orders of magnitude increase of the carrier life times and allowing us to estimate an ultra-low surface recombination of $S \sim 530$ cm/s, which, to our knowledge is a record for nano structures. Most importantly, this ultra-low surface recombination is a key enabler for the future development of efficient, high power, and room-temperature electrically pumped nanocavity lasers.

2. Fabrication and Technology

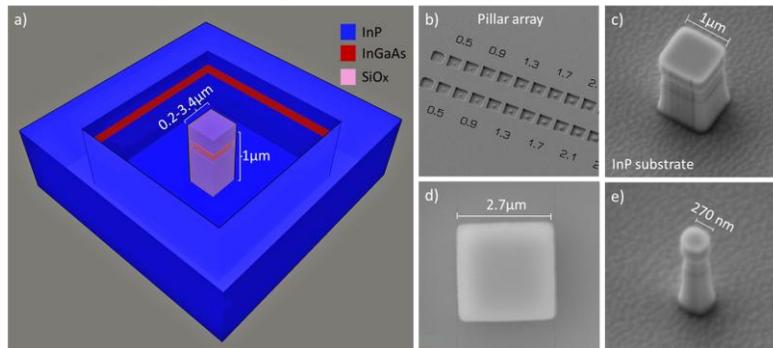
A series of experiments were performed to test (NH₄)₂S (ammonium sulfide) plus thin SiO_x capping passivation on undoped InP/InGaAs/InP nano-pillars. The layer stack is a *pn*-junction of InP with bulk InGaAs (350 nm thickness) material as shown in Figure 1 a). The sample comprises 7 equal patterns of two rows; each with 15 square pillars. The pillars side ranged from 270 nm to 3.4 μm.

The fabrication involved electron beam lithography (EBL), followed by dry etching with inductively coupled plasma (ICP) to etch the pillars until ~1 μm depth. It included wet etching steps plus oxygen plasma treatment to clean the surface of the pillars. Figure 1a-d) shows the schematic of the pillar after being capped by SiO_x and

Scanning Electron Microscope (SEM) pictures of the pillar array, a 1 μm cross-section side pillar, the top view of a pillar with 2.7 μm side and the smallest test pillar with 270 nm side.

The samples, named R1-Q1 to R1-Q7 were tested with two different passivation approaches: both processes started with a standard oxygen plasma cleaning of 10 minutes at 300W plus 2 minutes into diluted H_3PO_4 followed by 10 s gentle etching of the active material done by $\text{H}_2\text{O}:\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2$ (5000:1:8). After that, the samples were divided for different tests as Table 1 in Figure 2a) shows. The first test comprised 5 cycles of oxygen plasma plus wet chemical cleaning of 2 minutes with H_3PO_4 followed by rinsing with ultra-pure water (UPW) and drying with an N_2 pistol. The second test implied ammonium sulfide as passivation agent where the samples were submerged in a solution of 100:10 $\text{H}_2\text{O}:(\text{NH}_4)_2\text{S}$ -20% solution during 5 minutes [3, 4].

For each test a thin cap layer of 50 nm SiO_x , or 10 nm Al_2O_3 was deposited on top of the pillars to obtain a lasting passivation. The deposition was done either with Plasma Enhanced Chemical Vapor Deposition (PECVD) at 300 C or Atomic Layer Deposition (ALD) at 150 C (see figure 2 a)), the deposition was performed right after the oxygen plasma or ammonium sulfide passivation processes.



nanopillars using a pulsed laser at 968 nm with a periodic pulse train at 10 MHz and pulse width <100 ps. The photoluminescence was coupled into a single mode optical fiber using a microscope objective and then guided to a superconducting single photon detector (SSPD). For the TR measurements, a histogram of photon arrival times was built by correlating the SSPD output with the pulse pattern generator trigger with a correlation card (Pico Harp 300). As expected, sample R1-Q5 shows longer carrier lifetimes for all pillars sizes (from 0.27 to 3.4 μm). Figure 3a) shows the lifetime comparison of a sub-micron cross-section pillar of 270 nm between the untreated sample (black), and the ones passivated with $(\text{NH}_4)_2\text{S}$ (blue) and $(\text{NH}_4)_2\text{S} + \text{SiO}_x$ (red). Impressive lifetimes of 24 ns (and 46 ns for 570 nm side pillar) were recorded. It has been demonstrated elsewhere that the lifetime of bulk InGaAs can achieve values of even 100 ns [5]. The implication of such long lifetimes is an extreme reduction of the surface recombination of the pillars. The surface recombination was estimated by performing a linear fit of the carrier lifetimes using the equation $1/\tau_{PL} = 1/\tau_b + 2S/d$ [2], where τ_{PL} is the effective carrier lifetime, τ_b is the carrier lifetime in the bulk material, S is the surface velocity, and d is the side length of the pillar with square cross-section. Figure 3b) shows the experimental data with sub-micron pillars and the linear fit from which the surface recombination is extracted. An ultra-low surface recombination of around 530 cm/s is estimated, which is to our knowledge a record low surface recombination for deeply etched sub-micron pn -junctions. This value is two orders of magnitude lower than the surface recombination velocity for the un-passivated samples, estimated to be $\sim 1.2 \times 10^4$ cm/s.

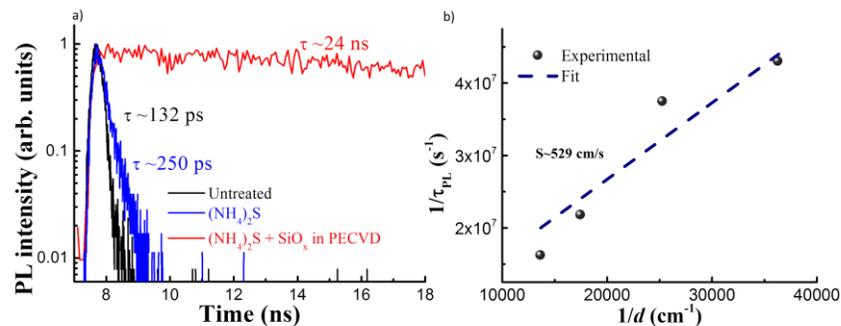


Figure 3 a) TRPL for a 270 nm side length pillar: untreated, with $(\text{NH}_4)_2\text{S}$ and with $(\text{NH}_4)_2\text{S} + \text{SiO}_x$, b) Surface recombination velocity of sub-micron pillars with $(\text{NH}_4)_2\text{S} + \text{SiO}_x$ treatment.

4. Conclusions

We tested two different passivation processes with a series of cycles of oxygen plasma and ammonium sulfide plus a thin capping of SiO_x or Al_2O_3 . We demonstrated a robust passivation process with $(\text{NH}_4)_2\text{S}$ + thin SiO_x capping. The passivation process is lasting and reliable. To prove it, we measured samples again after some days and retrieved the same excellent values. We obtained from the integrated μPL measurements around 100x better results in comparison with samples without passivation and TRPL that resulted in impressive carrier lifetimes of 46 ns for sub-micron side pillars, a value close to the bulk material. With the long lifetimes we were able to estimate an ultra-low surface recombination value of 530 cm/s; which, to the best of our knowledge, is a record low value of surface recombination velocity for deeply etched III-V nano pn -junctions. These results are highly encouraging for the fabrication of electrically pumped sub- μm lasers or any active deeply etched sub- μm device.

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