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Silicon Nanocrystal Laser

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Auspices Statement

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FY04 LDRD Final Report
Silicon Nanocrystal Laser
LDRD Project Tracking Code: 03-FS-010
June Yu, Principal Investigator

Abstract

The purpose of this feasibility study project was to attempt to demonstrate the silicon-nanocrystal-based laser. Such a silicon laser (made using conventional silicon-manufacturing technologies) would provide the crucial missing link that would enable a completely-silicon-based photonic system. We prepared thin layers of silicon nanocrystal material by ion-implanting Si in fused silica substrates, followed by a high temperature anneal process. These Si nanocrystals produced intense photoluminescence when optically pumped with ultraviolet light. Laser structures based on Fabry-Perot cavity and distributed feedback (DFB) designs were fabricated using the Si nanocrystals as the "lasing" medium. We optically pumped the samples with CW lasers at 413nm wavelength to quickly assess the feasibility of making lasers out of the Nanocrystal Si material and to verify the gain coefficients reported by other research groups.

Introduction/Background

Bulk silicon, the basis of our telecommunications and computational infrastructure, possesses an indirect band gap, and therefore, does not exhibit room-temperature luminescence. It is superbly used for transistors and photodetectors, and when oxidized to form silica glass, as an excellent waveguide. It would be a wonderful thing to use it also for a photo-transmitter (i.e. laser or LED.) This would in principle enable phenomenal optical integration of all the functions of computing and communicating—transmitters, optical interconnects, amplifiers, fanouts, and other optoelectronic functions could be realized. So, the discovery [1] of bright deep-red luminescence from a special nanoscopic (amorphous) form of silicon was big news. The quest for luminescence and lasing (or at least, gain, the precursor of lasing) in Si has since been widely embraced, as evidenced by conference proceedings and edited volumes containing reports from workers at various distinguished institutions [2,3]. Reports [4] by an Italian group of significant optical gain in Si nanocrystals generated a lot of excitement in year 2000 and got reported in widely-read trade and science magazines. [5 - 8] Investigations by the Pavesi group and others[9] have showed the ability of such Si nanocrystals to act as an optical gain media with gain coefficients reported as high as 100 cm^{-1}. The status of Si nano-material gain measurements as of roughly year 2002 is described in a book [10] containing a chapter "Optical gain from silicon nanocrystals" by Albert Polman, critically reviewing the work of Pavesi et al. [5] .

Research Activities

To quickly assess the feasibility of making lasers out of the Nanocrystal Si material, we fabricated a series of Fabry-Perot structures incorporating the Nanocrystal Si material. The Si nanocrystals were made by ion implant Si atoms into fused silica substrates. The ion implantation steps were performed at Oak Ridge National Lab's SMAC facility. A number of different recipes, consisting of differing concentrations of silicon atoms, differing annealing temperatures, differing annealing durations, and differing implantation energies were tried. The recipe that produced the largest photolumincescent characteristics is the one we used to fabricate our Fabry-Perot and DFB samples. The chosen ion implantation recipe gave a relatively uniform concentration profile of 5E21 Si atoms/cm3 throughout the top 500 nm of the fused silica substrate. The Si ion implanted samples were annealed for one hour at a temperature of 1100 degrees Celsius. Annealing the samples in a passivating atmosphere (usually of O_2 and/or H_2) allows the excess Si atoms to condense into "nanoclusters" or "nanocrystals" or "quantum dots" containing hundreds of atoms. The nanocrystals are a few nm in size and hence, exhibit quantum effects. Notably, the symmetry that caused bulk Si to have an indirect band gap is broken, allowing optical emission to occur. Furthermore, "particle-in-a-box" scaling causes the optical transition energies (wavelengths) to depend on particle size. [11, 12] Surface chemistry (i.e. the "recipe") decisively affects the optical properties as well. [13, 14] Because a finished sample is doped with high-refractive-index semiconductor particles, the doped layer has a refractive index greater than that of the neighboring bulk silica, automatically producing a waveguide (Figure 1 & 2.)

For our samples, the resulting nanocrystals have an estimated mean diameter of approximately 3 nanometers as diagnosed by TEM. A number of Fabry-Perot samples were prepared. If the nanocrystal Si indeed has the net gain numbers reported by the Italian group, we expected to see lasing when the samples are sufficiently pumped.

We also generated samples by applying Bragg gratings to the Si implanted fused silica substrates, to serve as end-reflectors for lasers. These DFB structures (Figure 3), which are relatively easy to fabricate given our unique access to the required microfabrication capabilities, give us another effective means of interrogating the lasing potential of the Si nanocrystal material.

We pumped the samples with a 1watt CW laser at 351nm and 413nm wavelengths.

Results/Technical Outcome

A transmission spectrum (figure 4) of a Si nanocrystal sample shows the normal UV cutoff of fused silica, along with a UV absorption caused by the Si particles, whose absorption strengthens in the UV. In the visible range, where the absorption is not strong, we see ripples due to Fresnel reflections from the top and bottom of the thin (0.5 μm) doped layer. Figure 5 shows one of our fabricated Si nanocrystal waveguide samples on a 2cm diameter SiO_2 wafer and the strong photoluminescence observed when the sample is illuminated by a 1 watt CW laser having a wavelength of 413 nanometers. Photoluminescence spectra (fig 6) of our samples, at two different dopant levels, along with one of a Pavesi [5] sample, show a peak in the red.

Evidently, higher doping produces a redshift, as one would expect from larger clusters produced during the annealing process.

The DFB samples were pumped at ORNL with a pulsed 337 nm wavelength nitrogen laser with pulse durations of 800 picoseconds and pulse outputs of 2.8 micro-joules. In addition to the expected broad photoluminescence curve centered about 750nm, we observed a sharp peak at 835nm in the leakage emission coming out of the top surface of the BFD structure (figure 7). The 835nm emission suggests that the actual channel waveguide index of refraction is 1.75.

However, no lasing was readily observed with our samples when pumped with 413 nm CW light. This result may be consistent with the recently published article from the joint Rochester and Italian group where they observed no gain for CW pumping, but net gain of $50cm^{-1}$ were observed under high intensity pulsed excitation[15].

Exit Plan

A clean demonstration of optical gain in the Si nanocrystal material will be highly visible in the laser, optoelectronics, and nanoscience communities. This is clearly a high risk, but high pay-off project.

Given the fresh results from the joint Rochester and Italian group, [15] we plan to conduct a complete set of laser gain measurements to fully characterize our Si nanocrystal material and waveguide structure with a tripled Nd:YAG laser. We will also have a try at the "variable stripe length" experiment, in hopes of getting results that can be interpreted. It makes a lot of sense to compare our findings with those of other groups. There are many resources at LLNL that could be engaged, since there is already a community of interested parties. [16 – 21] Microscopy, chemical analysis, etc. would be helpful in our project, which really ought at that point to be considered "materials science." Since we have world-class grating-fabrication capabilities (e.g. for NIF,) we would want to try different kinds of feedback and coupling structures.

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Figure 1 Nanocrystal Si waveguide

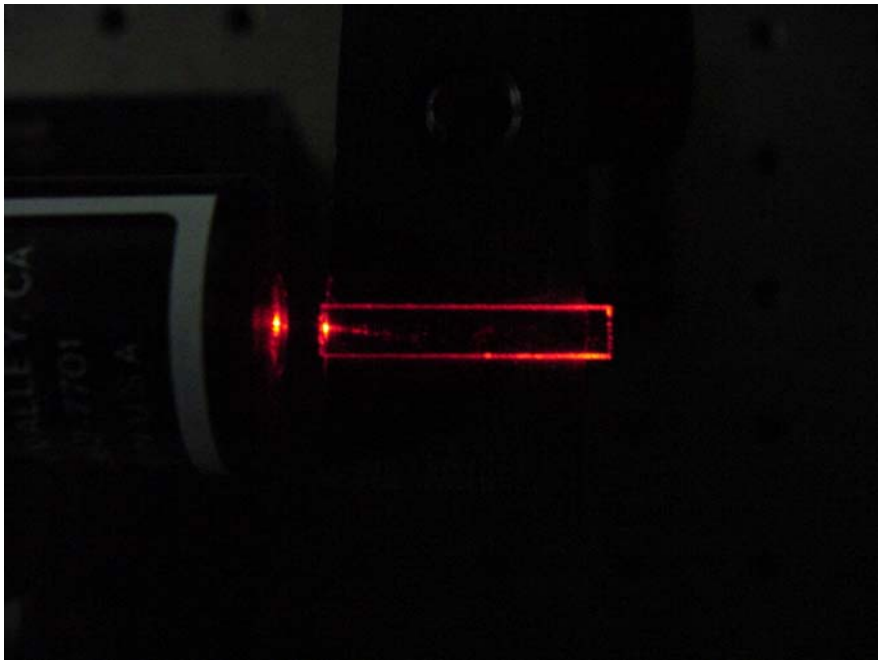

Figure 2 Photo showing wave guiding in the Fabry-Perot Silicon Nanocrystal sample

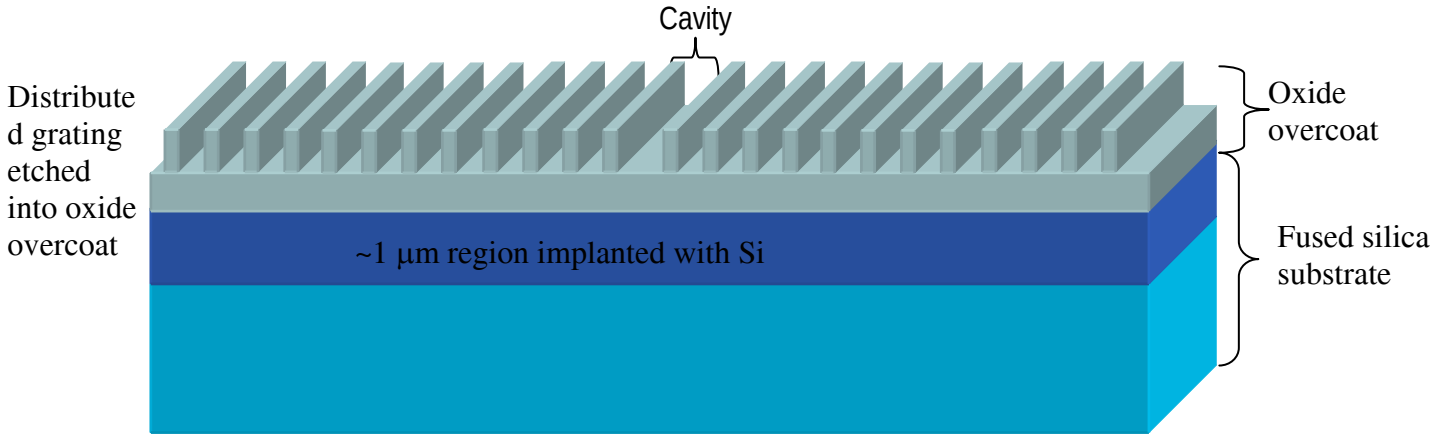

Figure 3a Schematic of DFB grating etched into the Oxide layer deposited over the Nanocrytal Si implant region.

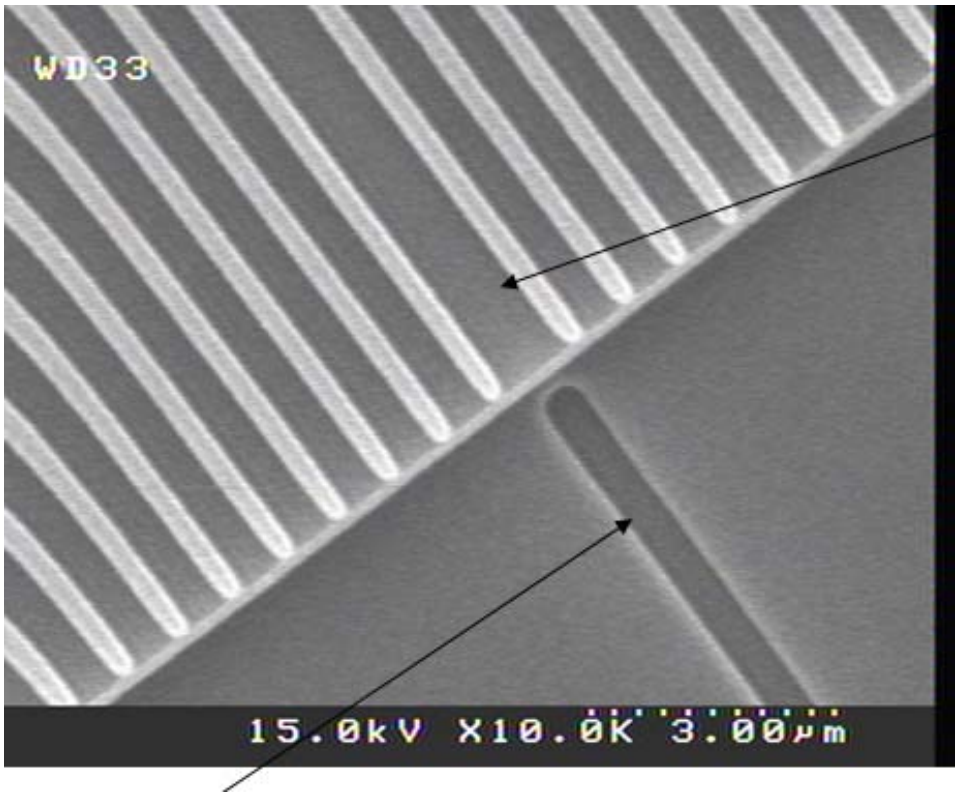

Figure 3b Glass slide patterned with distributed feedback focus exposure matrix

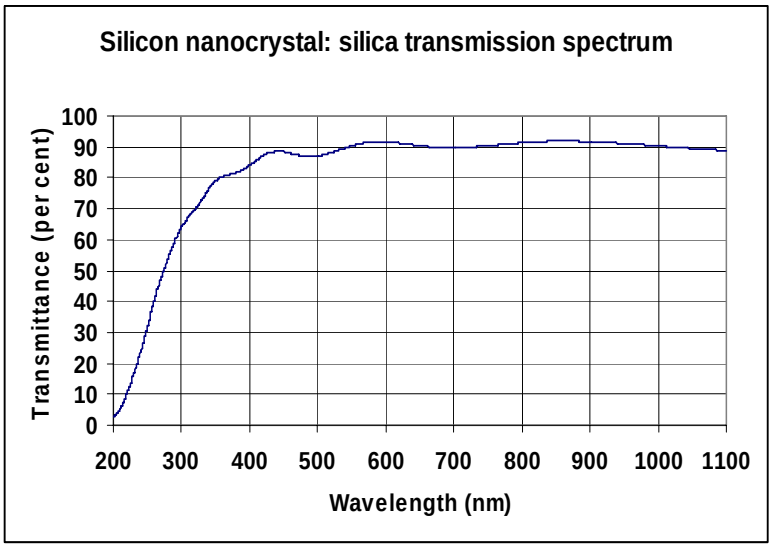

Figure 4 Transmission spectrum of ion-implanted, annealed sample

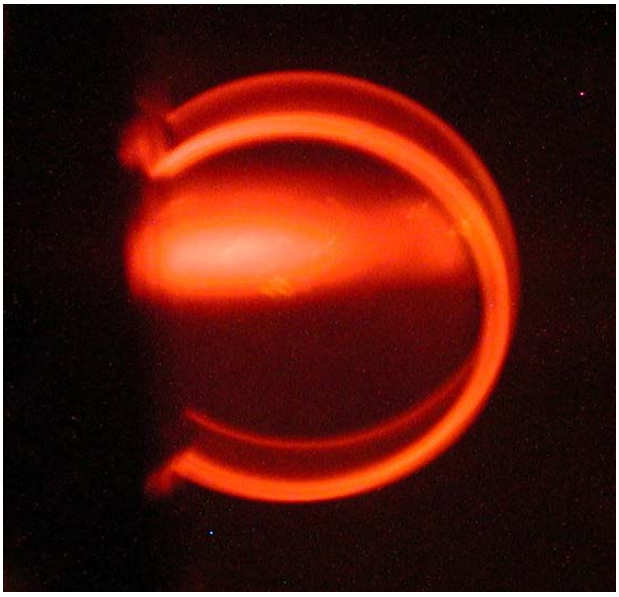

Figure 5 CCD image showing strong photoluminence from Si nanocrystals embedded in 2cm diameter SiO2 wafer. Pump light is from a 1watt CW 413nm Krypton ion laser.

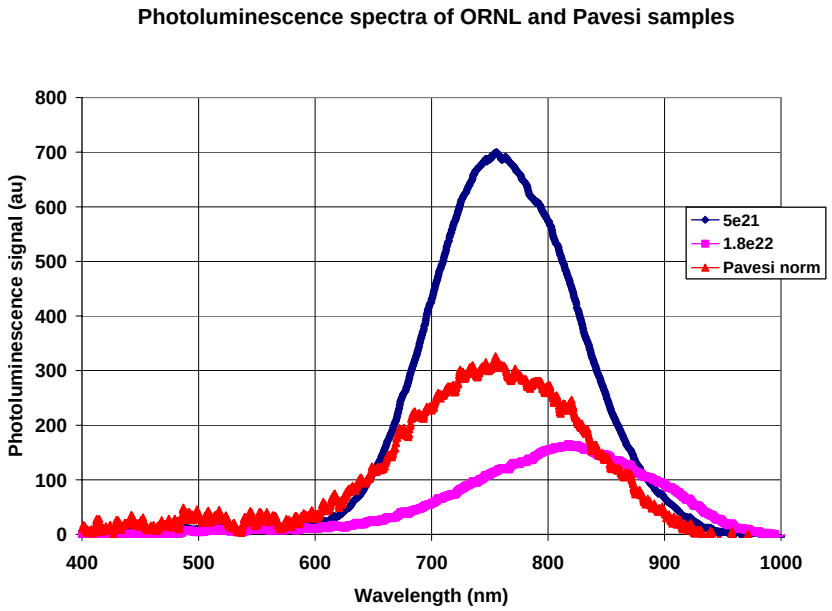

Figure 6 Photoluminescence spectra showing red emission from Si nanoclusters

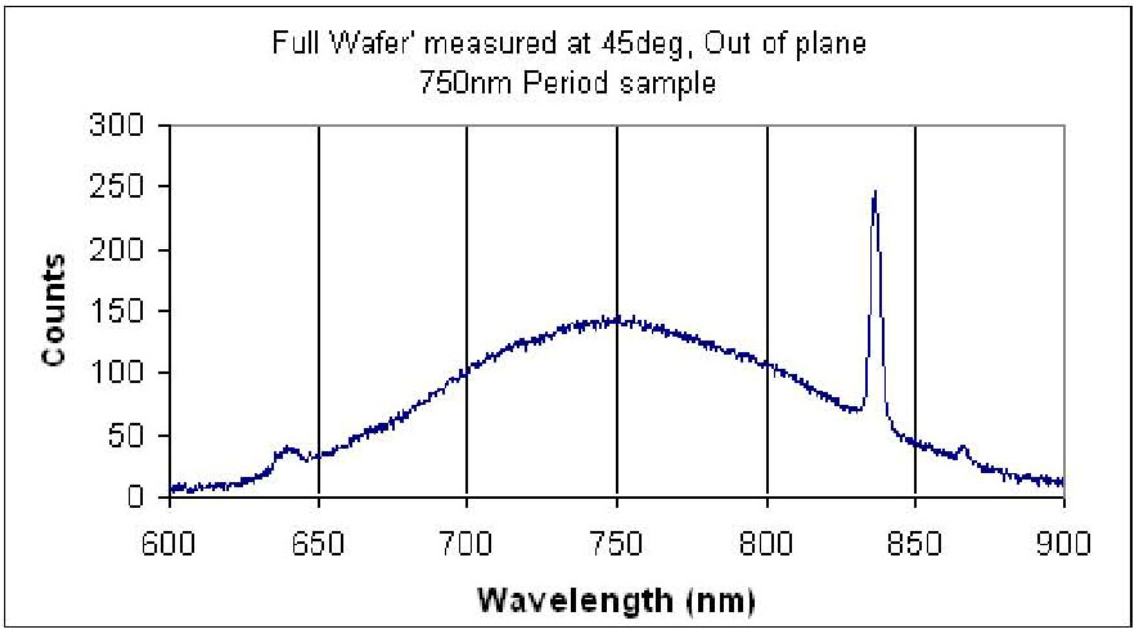

Figure 7 Spectroal emission shwoing the broad photoluminescent spectrum and the narrow distributive feedback emission at 835nm