Do Silicon Nanoparticles Exhibit Optical Gain?

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Abstract:

Room temperature photo-luminescent (PL) silicon nanoparticles (Si-nps) have been shown to emit across the visible light spectrum (400-800 nm) and have been used in light emitting diodes (LEDs). These same particles may be used as the active medium in a laser if they exhibit optical gain. This research focuses on determining if these particles produce net stimulated emission by utilizing a frequency doubled titanium-sapphire laser in a pump-probe configuration. With a pump wavelength of 400 nm and a probe of white light incident on a solution of hydrosilylated Si-nps in toluene, absorption across spectrum dominates any stimulated emission present.

Introduction/Background:

The focus of this research is whether photo-luminescent (PL) silicon nanoparticles (Si-nps) exhibit enough optical gain to produce a lasing medium. The Si-nps tested are created through silane (SiH_4) plasma by Dr. Stephen Campbell's and Dr. Uwe Kortshagen's groups, both of the University of Minnesota. Unlike bulk silicon that does not emit light because of its indirect band gap, these particles emit across the visible spectrum when optically excited [1]. The emission spectrum is dependent on the particle's size: the smallest particles at 3 nm emit blue light while larger particles, 5-6 nm, emit red light [1]. The size of the particle is determined by the residence time within the initial plasma and whether or not there is an etching plasma. The residence time is altered by changing the flow rates of the SiH_4 and Argon (Ar) in the initial plasma and etching requires a sulfur hexafluoride (SF_6) plasma [1]. As the wavelength decreases so does the quantum yield (QY): deep red (> 800 nm) QYs of 70% have been realized and blue QYs rarely reach 5% [1,2]. These particles have been used in light emitting diodes (LEDs) but because of the poor electro-luminescence of the Si-nps efficiencies are < 2% [3,4]. Because these Si-nps have high quantum efficiencies they are proposed to exhibit enough optical gain to allow an optically pumped laser to be produced.

Experimental Methods:

To test for the optical gain of these Si-nps, a solution of the particles was opted for testing procedures. Suspending the Si-nps in toluene required the particles to be hydrosilylated which is accomplished with the use of the ligand dodecene [2]; this process replaces the hydrogen terminated surface of the Si-nps with carbon disallowing the particles to aggregate. The dried, oxygen-free hydrosilylated particles were then suspended in toluene with a necessary optical density of approximately 0.2dB in a 1 mm static cell at 400 nm.

Several samples of particles were utilized to determine the concentration of Si-nps that was necessary to achieve a 0.2 dB optical density which would provide adequate signal. Two milliliters of solution were initially produced by using a 1 mL syringe; this solution had a concentration between 8 and 10 mg/mL. One mL was removed from this oxygen free vial and placed in a second non-evacuated vial. The original vial's concentration was then halved by injecting 1 mL of toluene in the vial and then sonicating the solution for two minutes. Another milliliter was removed and placed in a third vial for testing; this process was performed until a concentration of approximately 1 mg/mL was achieved. These different concentrations were then tested for their absorption characteristics in an Olis Cary 14 spectrophotometer. Figure 1 illustrates the absorption characteristics of different concentrations of Si-nps. From this information a concentration of approximately 2 mg/mL was chosen for testing. (See Figure 1.)

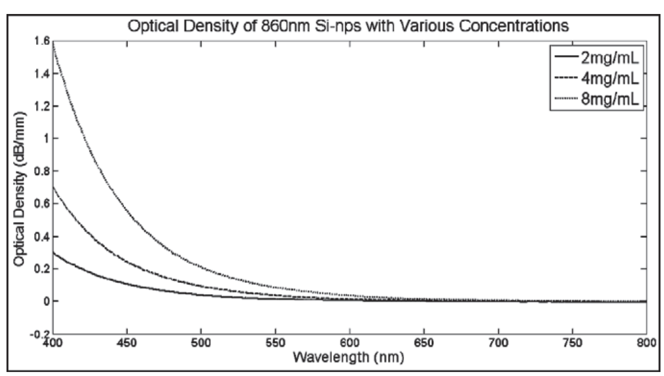

Figure 1: Optical density data for Si-nps.

The correct concentration colloid was then tested using a frequency doubled titanium-sapphire laser. The pulsed laser was incident on a static cell with a thickness of 1 mm which housed the colloid; 400 nm light was utilized for the pump and white light was used for the probe. Time delays of 500 fs, 1 ps, 10 ps, 50 ps and 500 ps were tested at five minute collecting times and a control delay of -5 ps was used.

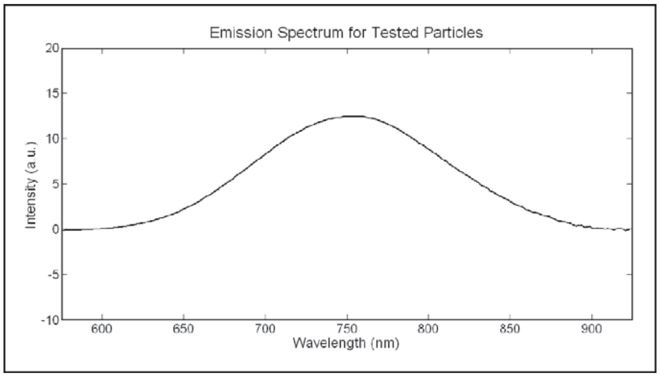

Figure 2: Spontaneous emission spectrum.

Results and Analysis:

Figure 2 summarizes the spontaneous emission of the tested particles showing a peak wavelength of 750 nm and a QY of 8%; this data was obtained with the use of an integrating sphere. Figure 3 illustrates the results of a representative test performed and depicts changes in the optical density—positive numbers exhibiting net absorption and negative values depict emission. Absorption dominates any emission present which is apparent because all the values are positive. Stimulated emission is expected at the wavelength of spontaneous emission which can be observed in the reduction of the absorption around 750 nm. Although stimulated emission is present in these Si-nps the emission cross section is dominated by absorption.

Conclusions and Future Work:

From the experiments performed thus far on the Si-nps, it appears optical gain is not observable with the current particles—implying laser applications are not feasible. If the quantum yield can be increased or stronger population inversion can be obtained there may still be a possibility of getting net emission which allows for future work.

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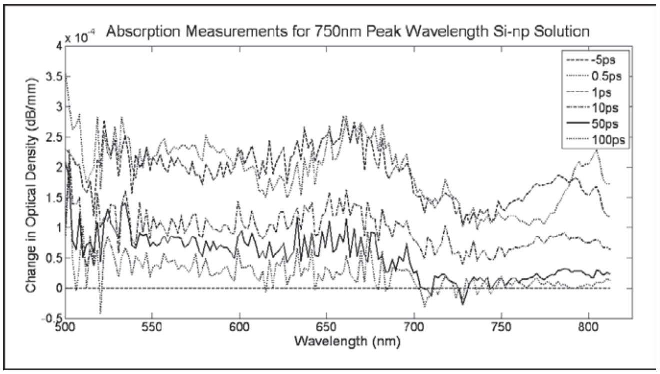

Figure 3: Pump-probe analysis of Si-nps.