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Light Emission Enhancement of Silicon Nanocrystals by Gold Layer

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Abstract**—**A thin gold metal layer was deposited on the top of silicon oxide films containing embedded Si nanocrystals (Si-nc). The sample was annealed in a gas containing nitrogen, and subsequently characterized by photoluminescence. We obtained 3-fold enhancement of photon emission from the Si-nc embedded in silicon dioxide covered with a Gold layer as compared with an uncovered sample. We attribute this enhancement to the increase of the spontaneous emission rate caused by the coupling of the Si-nc emitters with the surface plasmons (SP). The evolution of PL emission with laser irradiated time was also collected from covered samples, and compared to that from uncovered samples. In an uncovered sample, the PL intensity decreases with time, approximately with two decay constants. Although the decrease of the initial PL intensity associated with the increase of sample temperature under CW pumping is still observed in samples covered with a gold layer, this film significantly contributes to reduce the permanent deterioration of the PL intensity. The resistance to degradation of light-emitting silicon nanocrystals can be increased by SP coupling to suppress the permanent deterioration. Controlling the permanent photodeterioration can allow to perform a reliable optical gain measurement.

Keywords**—**Photodeterioration, Silicon Nanocrystals, Ion Implantation, Photoluminescence, Surface Plasmons.

I. Introduction

DURING the last decade, much attention has been focused on the investigation of the optical properties of Si nanocrystals (Si-ncs) incorporated into SiO_2 since these exhibit strong photoluminescence PL around 750 nm. This PL band is one of the most promising candidates for providing laser gain and thus realizing a Si based laser. [1], [2] It has been shown that Si-ncs have the ability to act as an optical gain medium with gain coefficients reported to be a wide range of gain values (from zero [3] up to $100cm^{-1}$ [4], [5]) as reported in the literature. Even using the same fabrication process to produce similar Si-ncs containing samples, experimental measurements still result in a broad range of gain values, [6] indicating that reliable and reproducible gain measurements are still lacking. It is well known that the optical gain measurements require that the sample be illuminated using relatively high average power laser, (continuous or pulsed laser systems) during a given acquisition time. On the other hand, we have recently observed that the photoluminescence (PL) emission exhibits a non negligible decay over time, even at low laser power intensities (~1 W/cm^2), [7] causing the optical gain measurements to be unreliable and irreproducible. Therefore, a sample treatment to circumvent the photodeterioration of Si nanocrystals embedded in SiO_2 samples would be highly desirable.

Several approaches have been recently tested [7] to increase the resistance to degradation of light-emitting Si-ncs embedded in dielectric layers. For example, UVC irradiation followed by annealing at an optimal temperature of 400 ^oC under nitrogen environment has been found to enhance the PL intensity by more than 100% and increases the resistance to degradation of light-emitting Si-ncs. [7], [8] In the present article, we investigate another possible sample treatment based on surface plasmon (SP) coupling technique induced by depositing a gold layer on silicon oxide containing silicon nanocrystals, followed by annealing; This treatment could be efficient to both increase the PL intensity and minimize the deterioration of PL emission of Si-ncs embedded in silicon oxide.

II. Experiment

The investigated samples were prepared by implanting Si^+ ions at energy of 50 keV into silicon oxide. Si^+ ion fluence of $5x10^{16}$ Si^+/cm^2 was used for this experiment. The expected depth profiles of the implanted Si were estimated using the SRIM code [9], showing a Gaussian distribution peaking approximately 70nm from the surface. The nucleation of Si-nc was activated by a one hour thermal annealing performed at 1100°C, under a N_2 atmosphere. A thin film of gold (200nm) over a chrome layer (10nm) was evaporated at room temperature followed by 1 hour annealing at 400^{o}C in N_2 atmosphere. Room temperature photoluminescence spectra were measured at various steps of the processing using a continuous 405nm (3.06 eV) diode laser focused to a 1mm diameter spot on the sample surface for an optical excitation intensity of 1.69 W cm^{-2}. A 550nm high-pass filter was inserted between the sample and the spectrometer to avoid detection of the excitation signal. The luminescence from the film was collected using an Ocean Optics QE65000 spectrometer, equipped with a CCD detector.

III. Results and Discussion

Si ion implantation into thermal SiO_2 formed on a silicon substrate and subsequent high temperature annealing (more than 1100°C) induce the formation of embedded luminescent Si nanocrystals by diffusion-controlled growth of Si nanoprecipitates. The peak energy of photoluminescence of the sample without the deposited gold film is approximately 1.61 eV for ion fluence of 5x1016 Si^+/cm^2.

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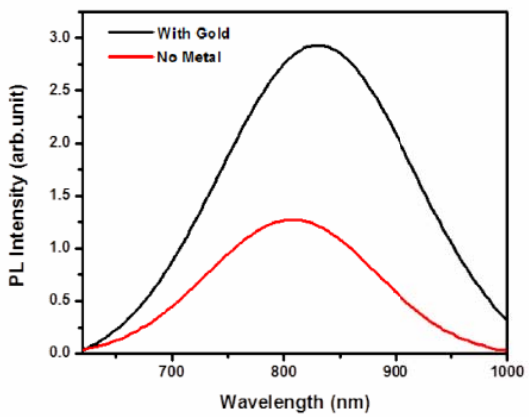

Fig. 1 PL spectra for Si nanocrystals in SiO_2 with gold and no metal layer

Fig. 1 shows PL spectra of uncovered and gold layer covered samples. The use of a gold layer has strongly enhanced the PL intensity from the Si-nc embedded in silicon oxide. Indeed, the PL peak of the treated sample with gold layer is 3 times that of untreated sample. This remarkable PL enhancement could be attributed to a strong interaction between the Si-nc emitters and the SP [10]. This resonant interaction might promote the spontaneous emission rates [11]-[14]. The SP coupling condition is dependent on the matching of energies between the SP frequency and the emission wavelength. The increase in PL intensity for treated samples is accompanied by a red spectral shift of 24nm. This feature suggests that the SP-coupling technique is more effective for wavelengths longer than 600nm for increasing the emission efficiency of materials. This behavior would be due to the fact that the SP resonance wavelength mode is coupled to the radiating dipoles in the nanocrystals, enhancing the intensity of the longer-wavelength portion of the Si-nc emission leading to a red shift of the PL spectrum.

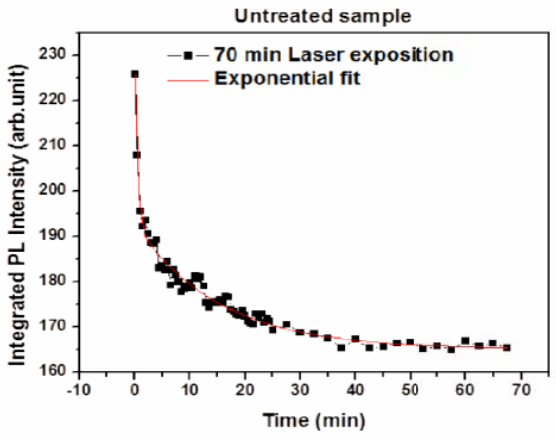

Fig. 2 Evolution of the spectrally integrated PL emission as a function of laser irradiation time

Fig. 2 shows the integrated PL emission as a function of irradiation time for the samples implanted at a fluence of $5x10^{16}$ Si^+/cm^2. There exists a significant decrease (or fatigue) of PL emission for successive laser exposures. After an irradiation time of 0.50min, the initial PL intensity has decreased of 15%. The intensity decreases has a 'fast' and a 'slow' decay component that can be fitted using a double exponential function, yielding two times constants. In our previous paper, we attributed the fast decay (t_1 ~ 0.5min) mainly to the local heating induced by the pumping laser. The second decay component of the PL emission is much slower (t_2 ~ 13.91min) and might be associated with the creation of defects at the Si-nc SiO_2 interface [15].

The time evolution of spectrally-integrated PL emission of samples covered with a metal layer is shown in Fig. 3. It is important to note that the initial PL intensities are greater by more than 250% than those of uncovered sample. The intensities of the PL spectra still decrease during the 70 min laser exposure. A glance at Figs. 2 and 3 informs us that the integrated PL intensity decrease is quite similar (~28%). However, the shape of the curves is different - the signal decrease being significantly faster for the treated sample. The results have been fitted to a two-time scale exponential decay. As inferred from the untreated sample, there is a fast decay (t_1 ~ 0.36min), mainly due to the local heating induced by the pump laser, and a slower time constant (t_2 ~ 14.98min for gold). It seems also that the slow decay time constant has increased for gold treatment suggesting that the SP coupling technique increases their resistance to damage induced by the laser irradiation and becomes more resistant to photodegradation. It is worth mentioning that new samples were used for each series of experiments.

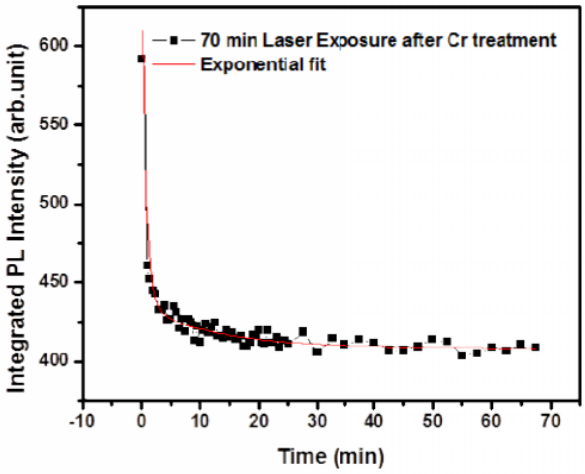

Fig. 3 Evolution of the spectrally integrated PL emission as a function of laser irradiation time after gold deposition and annealing treatment

IV. Conclusion

In summary, we have investigated the effect of depositing a metal layer on Si-nc embedded SiO_2 samples. A strong enhancement of the PL intensity has been was observed, which is due to an increase of the spontaneous emission rate

caused by the coupling of the Si-nc emitters with the surface plasmons (SP). We have also observed a favorable effect of the metal layer on the degradation of Si-nc PL emission: a significant reduction in the damage induced in the Si-nc by laser excitation. We have pointed out the benefits of SP coupling, which is a very powerful method to enhance light emission efficiencies of Si-nc, to increase the resistance to degradation of light-emitting Si-nc. This technique is very simple, easy to carry out, and can be applied to various materials that suffer from low quantum efficiencies (as reported by Okamoto et al. [10]-[13]) and photodeterioration. Following this treatment, a reliable gain measurement can be performed immediately after the first PL signal decay (fast time constant) associated with the local heating.

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