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NONLINEAR REFRACTIVE INDEX OF SILICON NANOSTRUCTURE PRODUCED BY PHOTO-ELECTROCHEMICAL ETCHING

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ABSTRACT

Self-phase modulated optical fringe pattern are used to study the nonlinear optical response of nanocrystalline silicon produced by photoelectron chemical etching. Irradiation time-dependent changes in the refractive index are calculated for various sizes of nanocrystallites. Fabrication of porous silicon contains silicon nano- structures has been carried out via process PECE on n-type Si wafer with >100< orientation and electrolyte solution contain Hydrofluoric acid HF concentrations of (25% HF), various laser wavelengths (532nmand442nm). The observed morphological changes using scanning electron microscopy reveals formation of silicon nanostructure. There is a strong correlation between the nonlinear optical phenomenon of silicon nanocrystallites and the nanocrystallites size distribution.

KEYWORDS: Porous Silicon, Nonlinear Phenomena, Refractive Index, Self-phase Modulation.

INTRODUCTION

Nonlinear optical properties of the nanoscale semiconductors have been a topic of fundamental interest and potential applications in switching devices [1-5]. An enhancement in the third order susceptibility and the change in the refractive index due to quantum confinement effect have been reported for laser ablated Si nanoclusters [6,7] and free-standing porous silicon materials[8] and silicon nanocrystals grown by plasma-enhanced chemical vapor deposition [9]. Many authors [10-13] have utilized the Z-scan technique to measure □3 when an incident photon has energy below the absorption edge of the nanoparticles, i.e. optical nonlinearity results from the anharmonic motion of bound electrons. Whereas other groups [14-17] have also utilized the self-phase modulation (SPM) of continuous wave (CW) laser beams to generate optical fringes in a nonlinear medium. The optical properties such as nonlinearity associated with changes in refractive index are significantly affected by the presence of nanocrystallites and size distribution of nanocrystallites [8,18]. This can lead to well- known phenomena of self- focusing and self-phase modulation [19]. Cotter et al [13] have reported the third-order optical nonlinearity of nanometer-size semiconductors caused by electronic quantum confinement.

Moreover, Koker and Kolasinski observed optical fringes during the laser-induced etching (LIE) process and attributed the fringe patterns formation to the optical interference and the Fresnel diffraction of the light reflected from the porous layer [20].

When an intense laser beam having a Gaussian profile, is incident on a medium containing nanocrystallites of Si, the refractive index of such material is altered by the intensity of the laser beam. As the nanocrystallites of silicon interact with spatially varying laser intensity, the refractive index of the medium changes thus the beam propagates in varied optical paths and hence spatial phase variation occurs. This leads to establish a visible optical fringe pattern in the transverse plane and the phenomenon is known as spatial self-phase modulation [21].

The far-field diffraction intensity distribution of the optical generated fringe pattern is given by the relation [17].

$$I(x) = I_0\left(\frac{2\pi}{\lambda Z}\right)^2 \left|\int_0^\infty r\,dr\,J_0\left(\frac{krx}{2Z}\right) \times \exp\left(-2\frac{r^2}{\omega^2} - i\phi(r)\right)\right|^2 \text{..................} (1)$$

Where I(x) is the far field diffraction of the Gaussian beam, Jo (x) = zero order Bessel function, k is the wave number in free space, Z the distance from the sample to the sample observation point, □ is the beam waste and □(r) is the phase factor. The change in the refractive index of the medium containing nanocrystallites of silicon across the propagation of the beam, □n(r), is written as [22,23].

$$\Delta n(r) = \gamma_L I(r)........(2)$$

Where γL is the nonlinear coefficient and I(r) is the incident laser intensity.
We propose that γL depends on the size distribution of the nanoparticles, thus can be written as

$$\gamma_L = \int_{L_1}^{L_2} f \frac{N(L)}{L} dL.......(3)$$

Where f is the coupling constant of light with the medium containing nanocrystallites and N(L) is the Gaussian distribution of nanocrystallites between L1 and L2, where L1 and L2 are the minimum and maximum sizes of the nanocrystallites.

In this paper, we have reported the optical fringe pattern of nanocrystallites during the laser etching of Si. The nanocrystallite size distribution is studied and analyzed using the self-phase modulation. Furthermore, the reconstructed surface morphology was studied using SEM, which revealed formation Si nanostructure, pits and different nanocrystallite sizes.

EXPERIMENTAL PROCEDURE

This process has more complicated set-up since it's a combination of laser induced etching process (LIE) and electrochemical etching process ECE. Among the etching methods available today, photo electrochemical etching (PECE) of the n-type silicon is studied particularly. It is a simple, low cost and controllable as well as its characteristics combine between features of photo electrochemical etching process and the laser-induced etching. Therefore, each type of silicon (n type) could be carried out in very short time using this method. When the sizes of the Nano crystallites are great we don't observe any optical fringe patterns. The laser intensity upon the Samples are used as parameters to assess the resultant nonlinear lensing result. Porous silicon layers were prepared using current density 2 mA/cm2 and an irradiation time of 7 minutes by wavelength of 532 nm upon a n-type silicon with resistivity (1- 10 Ω .cm). Sample was immersed in an HF acid of 25% concentration and absolute ethanol was additional for the aqueous solutions.

The set-up contained of a power supply as current source; ammeter as well as aqueous HF acid in Teflon cell with ethanol to prevent or minimize the hydrogen bubble through anodization. Careful design from the Photo electrochemical cell was required to obtain good uniformity for the porous structure in addition to the light source and the focusing lens, also to obtain corresponding interference optical fringes pattern. Fig (1) displays a schematic diagram from the PECE set-up. Quartz lens of about 10 cm focal lengths has been employed to collimate of the laser upon the front side from the Si surface through the Photo electrochemical etching method.

The current density used for PECE process of silicon was in the range of (2 mA/cm2). The process has been carried out at different etching time (2-5 min.). Then, samples were removed from the solution, rinsed in ethanol, and dried under a stream of air to avoid oxidation of the surface.

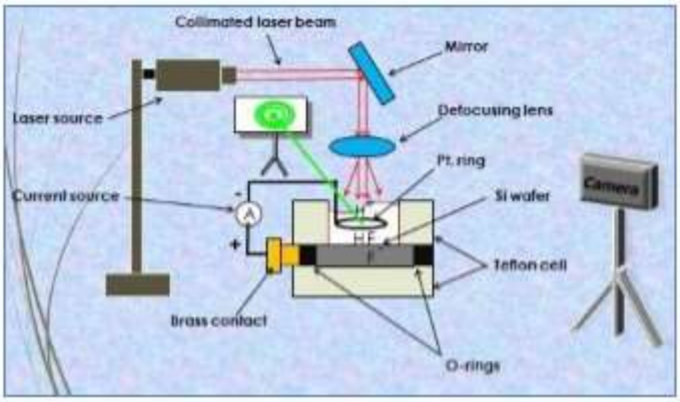

Fig (1): Schematic diagram of photo-electrochemical etching set-up.

RESULTS AND DISCUSSION

Nanocrystallite size distribution

Directly after leading the laser onto the crystals, the reflected beam has the intensity profile of the incident beams. A halo for diffusing light develops around the central spot which gradually separated from the central spot and forms a ring around it, leaving a dark circular progressively increases in diameters. A second light ring forms within the dark circle, which subsequently grow in diameter with times. It typically takes few minutes for a rings to transform from an indistinct halo which involved to the central spot to a distinct ring. The sequence is constant as subsequent ring form as showed.

Figure (2) illustrates the experimentally observed optical fringe patterns for different etching time in the right column and the corresponding theoretically calculated optical fringe patterns in the left column. We found that as the photoelectron chemical etching proceeds, number of fringe pattern increases and the mean nanocrystallite size decreases to reach 0.9 nm at 5 minutes as given in Table (1). The change in the refractive index (Δn) decreases with time.

It seems that the appearances of optical fringes are related with the size of the nano-Si. Fringes disappear when these sharper structures are etched away leaving shallow structures deep into the surface.

Appearance of optical fringes takes place again as a new layer of nano-Si structures appear due to etching of the already formed shallow structures deeper into the surface. When the mean size decrease the maximum change in refractive index increase as reveal in Table (1) which gives the calculated nanocrystallite sizes for different irradiation time and the experimentally measured number of fringes. Fig.(3) shows the relation between mean nano crystallite size and etching time.

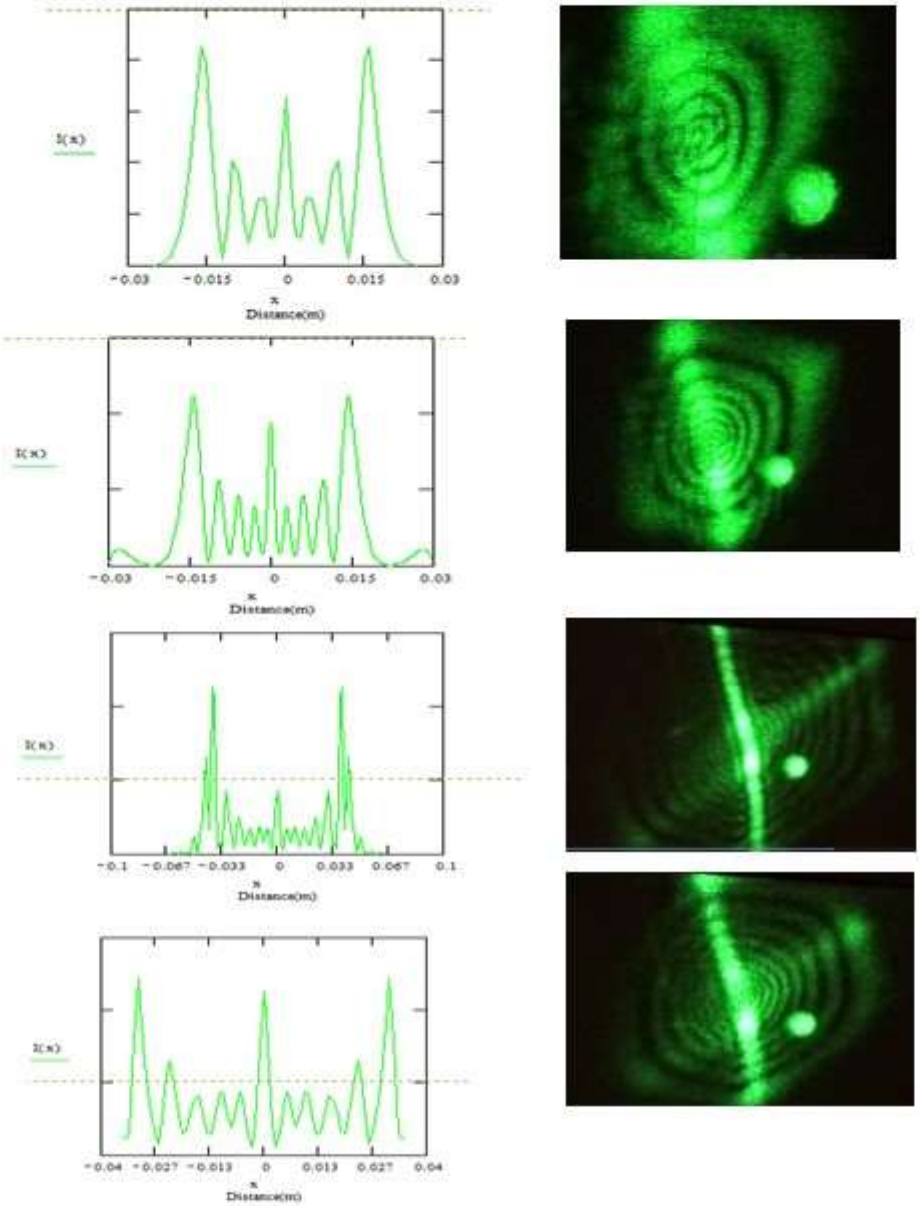

Fig. (2) manifestation of the laser beam reflected from the surface of the nano-Si prepared by PECE at an incident power (15mw) for different irradiation time and their theoretically calculated plot of optical fringe patterns(λ=532nm)

Table (1) gives the theoretical calculated nanocrystallite sizes for different irradiation times.

Time (mint)	No.of fringes	L1(nm)	L2(nm)	L0(nm)	Δn
2	**4**	**0.03**	**6**	**3.015**	**0.272**
3	**5**	**0.2**	**4.2**	**2.35**	**0.278**
4	**6**	**0.07**	**5.7**	**1.3**	**0.492**
5	**8**	**0.1**	**4.2**	**0.9**	**0.497**

Fig (3) The mean nanocrystallite size as a function of irradiation time.

Morphological Properties

The morphological aspects of the nano (PSi) surface like pore width, pore shape and silicon nano spacing between adjacent pores are a function to the experimental. The effects of experimental conditions like etching current density, etching time, type of silicon wafer were studied based on the surface morphology images. The meaning of pore refers to pit with large depth compare with its width. The pore initiated as a defect at the crystalline silicon surface. Any varying in parameter of laser, material interraction process such as illumination, wavelength and etching current density would lead to (PSi) layer process pores with various sizes, depths and shape. The etching process was achieved in various irradiation times at a constant light power density of about for n-type Si. One can easily distinguish two regions in the formation process of silicon nanocrystallites. The first region is observed at short etching time, in which disconnected pits with irregular structure on a silicon wafers are observed and these pits are commenced with the initiation of etching process on silicon wafers. Increasing the etching time will increase the number of these pits. The second region is observed at long etching time in which, the porous structure will be formed with different morphologies and depths on the silicon wafers. The first region is clearly observed in SEM micrograph as presented in figures (4) and (5).

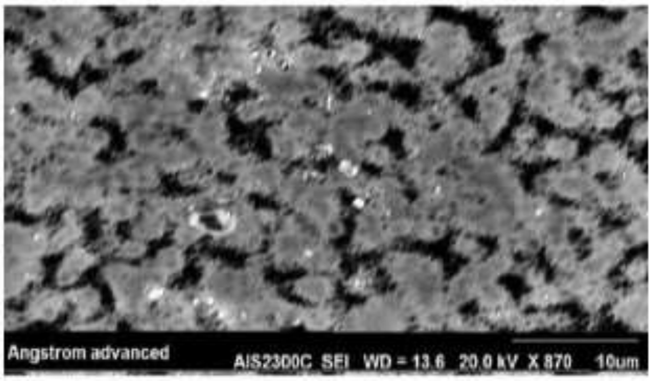

Fig.(4) SEM images of porous silicon sample prepared by green laser (532 nm), current density (2 mA/cm^2) acid concentration 25% and irradiation time 4 min.s

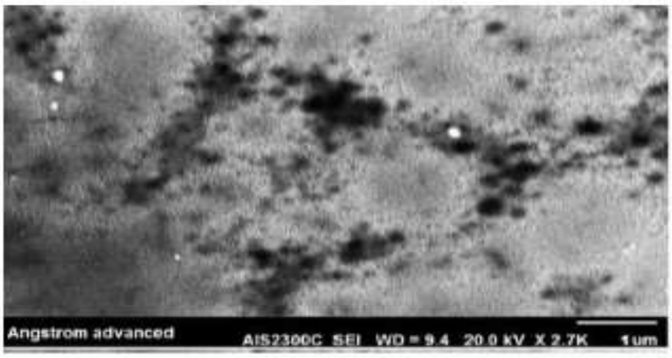

Fig.(5) SEM images of porous silicon sample prepared by green laser (532 nm), current density (2 mA/cm^2) acid concentration 25% and irradiation time 10 min.s

CONCLUSIONS

Formation of optical fringe patterns from nano-Si prepared by photo- electrochemical etching is investigated. The optical fringe patterns show the concentric optical fringe patterns are evolved from single bright spot as the irradiation time for etching is increased, in addition to that number of rings increases when the etching time is proceeding. The optical fringe patterns are found to be due to the self- phase modulation of light in spatial domain. A medium containing nano-Si materials would behave as a nonlinear medium under the influence of intense laser beam. Thus, a change in the refractive index of the porous silicon is expected which depends on the size of nano-Si. The SEM investigation revealed the formation of a pits-like structure

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