Third order nonlinear optical properties of nano-structured Si

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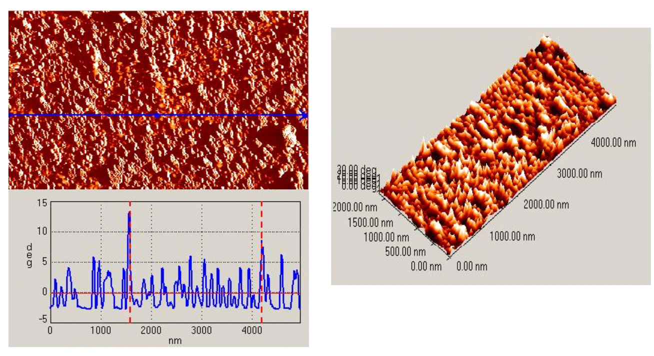
Nano-structured silicon (Si) is an interesting material for photonics, due to its small reflection, convenient processing and infiltration possibility (with other materials attractive for emission and nonlinear optical properties).

In this paper, we simplify and apply optical modeling for the Bruggeman-type nano-composites, to the calculation of the effective optical linear and nonlinear properties of nanostructured silicon. In our study, we gave a particular attention to nano-porous silicon samples (nPS).

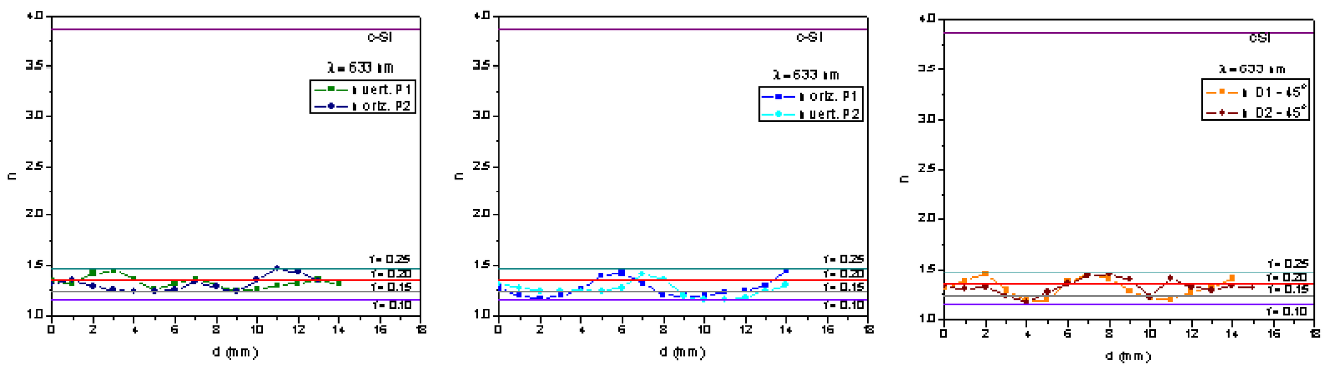
Structural and linear optical properties of nano-crystalline porous silicon

AFM images of nPS sample



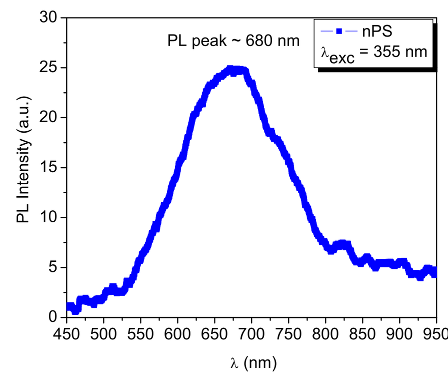
- Alveolar walls with thickness of ~ 100 nm
- Structure of the nPS sample surface.

Reflectivity spectra



- Experimental measurements of the nPS reflectivity provide a new optical method to measure the average volume fill fraction of Si (f_{Si}). In our case, f_{Si} = 0.18 and the average linear refractive index, n = 1.303

Photoluminescence spectrum



- Maximum emission energy (E_{PL}= 1.83 eV) of the excited nPS sample and its band-gap energy (E_g~ 2 eV).

The investigated nPS samples were obtained by electrochemical etching of bulk Si wafer in hydrofluoric acid (HF) [1,2,3].

Linear and nonlinear optical properties of nPS described by Bruggeman model

The properties of porous silicon can be described by Bruggeman geometry, because it consists of two randomly intermixed components and the dimensions of the Si walls is much smaller than the wavelength.

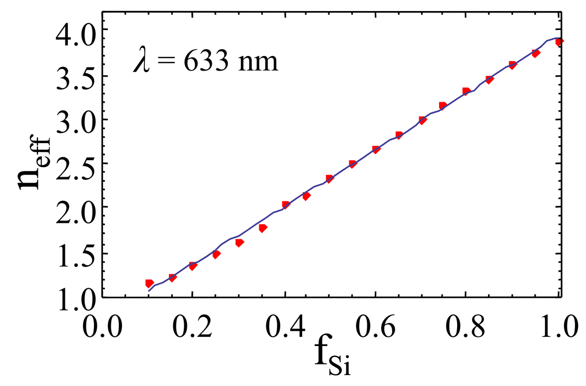
According to Bruggeman model, the effective linear dielectric constant of nPS layer can be described by the following equation [4,5]:

$$\varepsilon_{eff} = \frac{1}{4}\left[2 - 3f_{Si} + \varepsilon_{Si}\left(3f_{Si} - 1\right) + \sqrt{8\varepsilon_{Si} + 2 - 3f_{Si} + \varepsilon_{Si}\left(3f_{Si} - 1\right)}\right] \qquad (1)$$

where f_{Si} is the volume fill fraction of Si and ε_{eff} is its dielectric constant, respectively. Using Eq. (1), we found for nPS an approximative linear dependence for the effective linear refractive index $n_{eff} = \sqrt{\varepsilon_{eff}}$ (at λ = 633 nm):

$$n_{eff} = 3.19 \cdot f_{Si} + 0.74 \qquad (2)$$

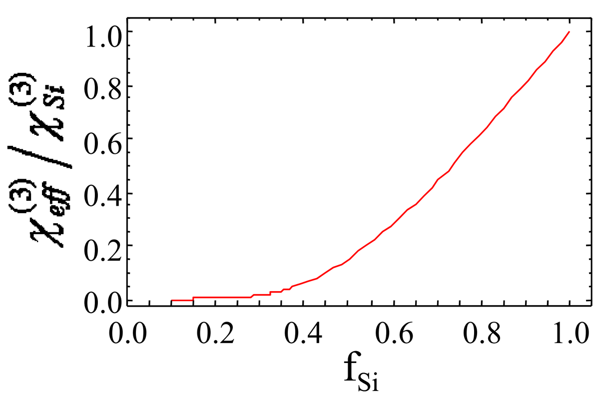
Dependence of n_{eff} of nPS versus f_{Si} for λ = 633 nm (the dots - calculated n_{eff} for different f_{Si}, the line - linear fit using Eq. (2))



The third order optical nonlinearity of nPS ($\chi_{eff}^{(3)}$) can be described using Bruggeman theory by:

$$\frac{\chi_{eff}^{(3)}}{\chi_{Si}^{(3)}} = \frac{1}{f_{Si}}\left(\frac{\partial\varepsilon_{eff}}{\partial\varepsilon_{Si}}\right)^2 = \frac{1}{f_{Si}}\left[\frac{1}{4}\left(3f_{Si} - 1 + \frac{2 - 9f_{Si}\left(f_{Si} - 1\right) + \varepsilon_{Si}\left(1 - 3f_{Si}\right)^2}{\sqrt{8\varepsilon_{Si} + \left(2 - 3f_{Si} + \varepsilon_{Si}\left(3f_{Si} - 1\right)\right)^2}}\right)\right]^2 \qquad (3)$$

Theoretical predictions of Bruggeman theory for $\chi_{eff}^{(3)}$ plotted as a function of f_{Si}



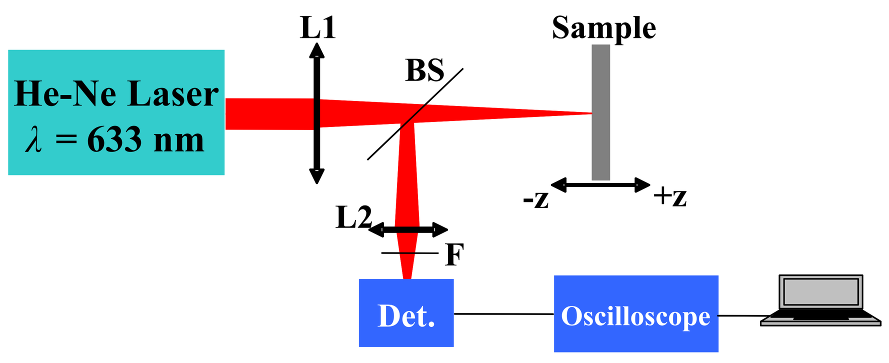
Using Eq. (3), we found a simple approximative quadratic dependence of the effective nonlinear third-order susceptibility on the Si volume fill fraction:

$$\frac{\chi_{eff}^{(3)}}{\chi_{Si}^{(3)}} \approx 1.46 \cdot f_{Si}^2 - 0.46 \cdot f_{Si} \qquad (4)$$

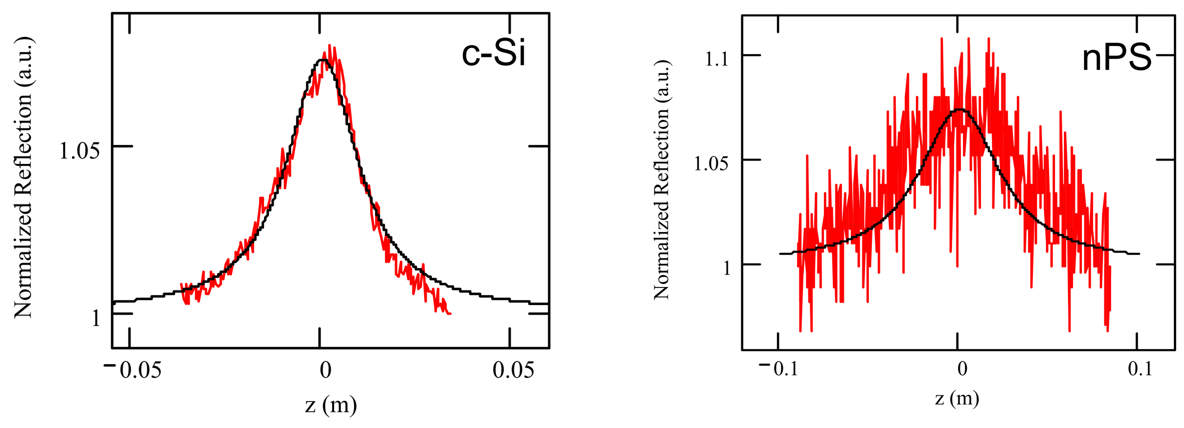
Third order nonlinearity experimental investigation by reflection Z-Scan method

Theoretical prediction for $\chi_{eff}^{(3)}$ obtained with Bruggeman model was experimentally verified by open-aperture reflection Z-Scan method [6]. In this method, the investigated sample is moving along the incident beam direction, passing through the focal plane of a focusing lens.

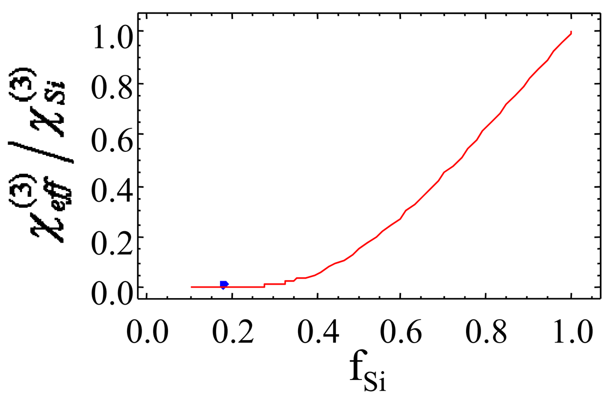
RZ-Scan experimental setup



RZ-Scan experimental results for c-Si and nPS



Theoretical predictions of $\chi_{eff}^{(3)}$ given by our Eq. (4)



Experimental measured optical nonlinearities, $\frac{\chi_{nPS\,eff}^{(3)}}{\chi_{Si}^{(3)}} = 1.3 \cdot 10^{-2}$, are in good agreement with the data obtained with our simplified model.

Conclusion

This study brings contributions for simplification of Bruggeman model for nano-composites, applies the adapted relations in the study of linear and nonlinear optical properties of the nPS samples and experimentally confirms the validity of these relations.

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