Strongly enhanced third order nonlinear response of periodically nano-structured silicon-on-insulator (SOI) measured by reflection Z-scan with femtosecond pulses

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1. Introduction

Third-order nonlinear optical response of SOI structures was recently investigated [1]. Third harmonic generation (THG) at excitation wavelengths of 800 nm and 1500 nm was studied in 1D SOI photonic crystals and in un-patterned SOI waveguide. The authors investigated TH reflectance, waveguide interference effects, and nonlinear Bragg diffraction in relation with the interplay of pump and TH beams propagation. A strong enhancement of the nonlinear conversion efficiency in TH was reported in 1D SOI photonic crystals in comparison with the efficiency observed in un-patterned SOI structures. On the other hand, the THG efficiency was enhanced in un-patterned SOI structure with respect to that obtained in bulk silicon [1]. These enhancements were explained by the field enhancement at the sidewall edges in 1D SOI, and by the vertical confinement of the exciting light in un-patterned SOI, respectively.

In the Z-scan methods, the investigated sample is moving along the incident beam direction, passing through the focal plane of a focusing lens. In the reflection Z-scan (RZ-scan) method, optical nonlinearities are quantitatively measured analyzing the beam reflected by the sample, instead of the beam transmitted through the sample, as in the transmission Z-scan (TZ-scan) [2, 3]. The RZ-scan method allows for the determination of the magnitude and the sign of the optical nonlinearity. In samples with nonlinear change of the refractive index, the reflection coefficient is dependent on the local beam intensity. In open aperture RZ-scan configuration, the refractive nonlinearities are measured by monitoring the total change of the reflection coefficient. These nonlinearities are responsible for the amplitude changes of the reflected radiation [3, 4-7]. In a closed aperture RZ-scan configuration, the absorptive nonlinearities, responsible for the phase changes in the reflected beam, are measured [6].

In this paper, we use the open aperture RZ-scan configuration, only. When the sample is far from the focal plane of the focusing lens, the intensity of the incident light is low and the reflection coefficient $R_0$ of the sample is not changing. Around the focal plane, the incident intensity is much higher and the nonlinear refraction occurs, changing the magnitude of the reflection coefficient. For a positive nonlinearity ($n_2 > 0$), the reflection coefficient $R(z)$ is increasing when the sample is approaching the focal plane, due to the increasing intensity, and it is decreasing after passing through the focal plane, where the intensity is decreasing, going down to the linear value, $R_0$. For a sample with a negative nonlinearity ($n_2 < 0$), the $R(z)$ dependence has a valley around the focal plane.

In a Z-scan experiment with high-repetition-rate fs pulses, the thermal nonlinearity, induced by cumulative heating effects, can play a significant role [8]. The use of a fast Z-scan device, as in our experiment, can decrease the thermal effects. In order to discriminate between electronic and thermal nonlinearities, we introduce a new procedure for reflection Z-scan, repeating the experiments with the laser in mode-locked or in c.w. regimes and maintaining the same conditions for the probes. Using this procedure, we can precisely measure both electronic and thermal nonlinear coefficients of nano-patterned SOI, un-patterned SOI and bulk silicon.
These results could be important in photonics for optical devices with large and fast nonlinear response [9] and with properties controlled by nano-patterning.

2. Experiment and discussion

The investigated sample is a silicon-on-insulator structure. The SOI wafer is produced by SOITEC by Smart Cut™ process [10]. It is composed of a silicon substrate, a SiO2 layer (2 µm thickness) and a thin Silicon layer (0.34 µm) on top. The conductivity of the Si top layer is around 10 Ω cm. By using standard electron-beam lithography and reactive-ion etching techniques [11], on the top Si layer was realized a bare periodic structure composed of 0.43 µm Si stripes separated by 0.07 µm air intervals (Fig. 1).

The refractive indices of Si and SiO2 at 800 nm are \( n_{\text{Si}} = 3.7 \), \( n_{\text{SiO2}} = 1.5 \), respectively, and the absorption coefficient of Si is, at this wavelength, \( \alpha \sim 1000 \text{ cm}^{-1} \).

![Fig. 1. A schematic of the periodic SOI structure.](image1)

The experimental setup used in the RZ-scan experiment is shown in the Fig. 2.

![Fig. 2. The experimental setup used in the RZ-scan experiment on SOI structures.](image2)

A Ti:Sapphire laser with wavelength \( \lambda = 800 \text{ nm} \), pulse duration \( \sim 75 \text{ fs} \), repetition rate 76 MHz and average power of 265 mW is used in the RZ-scan experiment. The laser radiation is focused with a 10-cm focal length lens (L) on the investigated sample, at normal incidence. The sample is fixed on an oscillating translation device which oscillates along the light direction (Z direction), with the movement adjusted in order to have the focal plane of the focusing lens in the middle of the Z-scan range. The beam reflected by the sample surface is extracted with the beamsplitter BS (a thick glass plate) and sent on a large area detector (Det) that allows collection of the entire beam reflected by BS (open aperture RZ-scan). To cut parasitic reflections the apertures D1 and D2 are used. An optical isolator made from a polarizer (P) and a \( \lambda/4 \) wave-plate (\( \lambda/4 \) WP) is used to prevent the laser un-mode-locking due to back-scattered reflections. The signal from the detector is visualized and stored for further processing by a two-channel digital oscilloscope (OSC).

The experimental RZ-scan curves obtained in nano-patterned SOI, un-patterned SOI and, for comparison, in bulk silicon are shown in the Fig. 3a-c.

![Fig. 3. Experimental RZ-scan curves obtained with the laser in CW regime (top continuous line) and mode-locked regime (bottom continuous line) and the theoretical fit (smooth dashed and dash-dotted lines) in nano-patterned SOI (a), un-patterned SOI (b) and bulk Si (c).](image3)
For a quantitative measurement of the refractive nonlinearities of SOI structure, the experimental RZ-scan dependencies were analyzed using the normalized reflected power given by the following relation [4, 6]:

\[
P(z) = 1 + 2 \text{Re}\left[R(n_2 + ik_2)\right] \int_0^\infty \left|E(\rho, z)\right|^2 \rho d\rho
\]

where \(\rho\) and \(z\) are the radial and axial coordinates, \(n_2\) is the nonlinear refraction coefficient, \(k_2\) is the nonlinear extinction coefficient, \(R = \Delta R/R_0\) is the relative change in the reflection coefficient and \(E(\rho, z)\) is the incident beam amplitude.

In the case of sample illumination at normal incidence and when \(n_0 \gg k_0\) (\(k_0\) is the linear extinction coefficient; in our case, for Si, \(n_0 = n_0 = 3.7, k_0 = k_0 = 0.006\)), Eq. (1) becomes:

\[
P(z) = 1 + \frac{4\Delta n}{n_0^2 - 1} \int_0^\infty \left|E(\rho, z)\right|^2 \rho d\rho,
\]

which shows that the amplitude of the normalized reflected power \(P(z)\) is proportional to the (small) change of the real part of refractive index, \(\Delta n\).

In the Z-scan experiments with high repetition fs pulses, the thermal nonlinearity, induced by cumulative heating effects, can play a significant role. The use of a fast Z-scan device in our experiment decreases, but cannot eliminate completely the thermal effects. In order to separate the contributions of the thermal nonlinearity and the electronic one from the overall nonlinear response of the samples, we have done Z-scan experiments with the laser working in the following two operation regimes.

1) In the continuous-wave (c.w.) regime, we can consider that the relatively small laser power produces only a thermal nonlinearity in the sample. The corresponding change of the refractive index can be written as:

\[
\Delta n_{\text{th}} = n_{2,\text{th}} I_0,
\]

where \(I_0\) is the laser beam intensity on the sample surface. In this case, from the RZ-scan curve, we can determine the nonlinear thermal coefficient \(n_{2,\text{th}}\).

2) In the mode-locked regime, the electronic nonlinearity becomes relevant in the sample, due to the very high peak power of femtosecond pulses:

\[
\Delta n_{\text{el}} = n_{2,\text{el}} I_p,
\]

where \(I_p\) is the peak light intensity of the fs light pulses.

In addition to this nonlinearity, the thermal one, produced by the heating of the sample with high-repetition-rate pulses, is present too. As the average power of the laser in the mode-locked regime is equal to the laser power in the c.w. regime, we can assume that the thermal effects produced in both regimes are equal:

\[
\Delta n_{\text{th}} = n_{2,\text{th}} I_{\text{av}} = n_{2,\text{th}} I_0,
\]

where \(I_{\text{av}}\) is the intensity on the sample surface, calculated with the average power of the laser. Due to the small nonlinear effects, the total change of the refractive index in the mode-locked regime can be written as a sum of the electronic and thermal changes:

\[
\Delta n = \Delta n_{\text{el}} + \Delta n_{\text{th}} = n_{2,\text{el}} I_p + n_{2,\text{th}} I_{\text{av}}.
\]

Our procedure to determine the electronic nonlinear coefficient, \(n_{2,\text{el}}\), from the normalized experimental RZ-scan curves is described below.

The RZ-scan curve obtained in c.w. regime is fitted with Eq. (2), in which the thermal refractive index change (3) is used and the value of the thermal nonlinear coefficient \(n_{2,\text{th}}\) is obtained.

The RZ-scan curve obtained in mode-locked regime is fitted with Eq. (2), in which the total refractive index change (6), as well as the value of \(n_{2,\text{th}}\) previously determined, are used. The result of this fitting leads to the determination of electronic nonlinear coefficient, \(n_{2,\text{el}}\).

Using this procedure to fit experimental RZ-scan curves in nano-patterned SOI, un-patterned SOI and bulk Si, it is possible to obtain both the thermal and electronic nonlinear coefficients of these structures. The results are shown in the Table 1.

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<tr>
<th>Table 1. The thermal and electronic nonlinear coefficients of nano-patterned and un-patterned SOI, compared with those of bulk silicon.</th>
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<tr>
<td>Nonlinear refractive coefficients</td>
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<tr>
<td>(n_{2,\text{th}} \times 10^{14} \text{ (m}^2\text{/W)})</td>
</tr>
<tr>
<td>(n_{2,\text{el}} \times 10^{20} \text{ (m}^2\text{/W)})</td>
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From this table one can see that the nano-patterning of SOI strongly enhances its nonlinear response. The electronic nonlinear response of nano-structured SOI sample is approx. 50 times stronger than the nonlinear response of un-patterned SOI, and approx. 125 times stronger than that of the bulk silicon.

3. Conclusions

We have measured precisely both electronic and thermal nonlinear coefficients of periodic nano-patterned SOI, un-patterned SOI and bulk silicon, using a reflection Z-scan setup with fs laser pulses (at wavelength of 800 nm) and a new procedure for discrimination between electronic and thermal nonlinearities.
We have observed a strong enhancement of third order nonlinear electronic response of nano-patterned (with periodic air channels of 70 nm width) and un-patterned SOI in comparison with that of bulk silicon: the electronic nonlinear response of nano-structured SOI sample is approx. 50 times stronger than the nonlinear response of un-patterned SOI and approx. 125 times stronger than that of the bulk silicon.

These results could be important in silicon photonics, for achievement of nonlinear optical devices with properties controlled by nano-patterning.

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