Origin of Waveguiding in Ultrashort Pulse Structured Silicon

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The origin of waveguiding in the bulk of silicon after sub-ps laser inscription is investigated. Locally resolved Raman measurements of waveguide cross sections and along the propagation axis reveal highly localized crystal deformations. These modifications consist of highly confined regions of silicon with a disturbed crystal structure accompanied with strain. This transformation is responsible for a local increase of the refractive index allowing localized waveguiding. On the basis of near-field measurements at an excitation wavelength of 1550 nm, the absolute value of the refractive index change is estimated to be in the range of $10^{-3}$.

1. Introduction

Silicon is the most important material in the semiconductor industry and provides the basis for modern optical and electro-optical technologies. One of the prime desires in communication industry today is the merging of electronic and photonic devices on the same chip. In order to achieve this goal, the direct writing of optical elements using ultrashort laser pulses is a promising approach. However, in contrast to glasses or other dielectrics, the direct inscription into silicon involves several difficulties, including strong spherical aberrations and pronounced nonlinear interaction.

Within the last two decades, numerous attempts have been performed to overcome these limitations. Recently, localized modifications in the bulk of silicon using ultrashort laser pulses have been demonstrated. This resulted in the first demonstration of waveguides written with fs laser pulses at a wavelength of 1550 nm by Pavlov et al. This year, we presented a detailed characterization of sub-ps written waveguides regarding near field distribution, scattering characteristics, and damping losses.

In this publication, we investigate the underlying mechanisms of ultrashort pulse written waveguides in silicon by means of Raman microscopy. The results obtained reveal highly localized defects within the crystal matrix accompanied with induced strain. These modifications are responsible for a positive refractive index change in the range of $10^{-3}$.

2. Sample Preparation

In order to investigate the underlying structural changes, numerous waveguides were written using the identical setup and method as presented in ref. [17]. The processing laser was an Er-doped fiber laser system (Raydiance, Inc.) delivering 800 fs pulses at a wavelength of 1552 nm. The pulse repetition rate was 400 kHz and the pulse energy 110 nJ. During waveguide writing, the laser beam was focused into the volume of mono-crystalline silicon (N/PH, (100), 1–10 $\Omega$ cm, 1 mm thickness) using a long-distance microscope objective (20 $\times$, NA = 0.4). The relative position between sample and objective was controlled using an automated 3D positioning system (Aerotech ANT 130). The writing direction was along the beam propagation axis by moving the focal region from the backside upward until approximately 200 µm below the front surface with a writing speed of 20 µm s$^{-1}$. After laser inscription, the back surface was polished. The characterization of the waveguides was performed in analogy to ref. [17] for measuring the supported mode-field distribution at 1550 nm.

The waveguides have a diameter cross section 3–5 µm (see Figure 1a) yielding a typical near-field intensity profile for a wavelength of 1550 nm as shown in Figure 1b. Based on the detected near-field profile, the underlying refractive index distribution was calculated by solving the Helmholtz equation. The corresponding result is shown in Figure 1c,d revealing a maximum refractive index change at the center of $2 \times 10^{-3}$. The order of magnitude of this refractive index change is comparable to fs written waveguides in glass.

3. Raman Analysis

In order to investigate the origin of the induced refractive index change, Raman microscopy was applied at a wavelength of 532 nm using a 100 $\times$ objective with NA = 0.85 (Renishaw inVia Raman microscope). The 2D mappings obtained were scanned with a step size of 0.25 µm in $x$- and $y$- direction, corresponding to the maximum available resolution of this system at 532 nm.

In the following, characteristic features of the Raman spectra are discussed. For a better overview, the discussed spectral ranges
Figure 1. a) Typical shadowgraphic image of a waveguide end facet illuminated from the back with a quartz tungsten-halogen lamp (Thorlabs, QTH10). b) Near-field intensity distribution of the same waveguide during excitation using a wavelength of 1550 nm, c) calculated refractive index distribution by solving the Helmholtz equation. d) Cut through the derived refractive index maximum along y and x axis. Processing parameters: pulse energy = 110 nJ, repetition rate = 400 kHz, writing speed = 20 µm s⁻¹.

Table 1. Overview on characteristic Raman signatures in silicon. Please note that related state corresponds to the crystal structure of silicon with the following abbreviations: a = amorphous, c = crystalline (diamond cubic); and relative amplitude refers to the positions given in Figure 2 (1/ref).

| Wave number [cm⁻¹] | Shape | State   | Relative amplitude | References |
|--------------------|-------|---------|--------------------|------------|
| 100–200            | hill  | a-Si    | 1.13               | [20–22]    |
| 300                | peak  | c-Si    | 0.97               | [22–24]    |
| 430–500            | peak  | a-Si    | 1.36               | [20,22,23] |
| 520                | peak  | c-Si    | 0.58               | [20,22,23] |
| 610                | peak  | c-Si    | 0.83               | [23]       |
| 930–1040           | plateau | c-Si | 0.71               | [20,23]    |

with corresponding crystal structures are listed in Table 1.[20–24] Important features for crystalline silicon (c-Si) are a sharp peak at a wave number of 520 cm⁻¹, as well as a broad peak between 930 and 1040 cm⁻¹. Moreover, smaller peaks at 300 and 610 cm⁻¹ are typical for silicon with a diamond cubic like structure (Si-I). In contrast to crystalline silicon, amorphous silicon (a-Si) shows a pronounced Raman signal between 100 and 200 cm⁻¹ and between 430 and 500 cm⁻¹. Furthermore, the c-Si main peak is absent. The transition from c-Si to a-Si is accompanied with a decrease of the crystalline features and an increase of the amorphous ones.[25,26]

Figure 2 shows the Raman measurements of the waveguide cross section obtained at the polished rear facet. In order to visualize the induced material modifications, the average Raman signal of the two spectral ranges 430–500 cm⁻¹ and 930–1040 cm⁻¹ are shown in Figure 2a,b. The waveguide area is clearly visible as a confined region with a diameter of 3–5 µm. The size obtained is in good agreement with the measurements presented in Figure 1 and [17]. Note that the diagonal lines are due to scratches from polishing. The overall Raman spectra for the two positions marked can be seen in Figure 2c. The reference signal representing the unmodified region was taken at the position marked with the white arrow in Figure 2a,b. This measurement shows the typical crystalline Raman features as expected (see Table 1). In contrast to this, the modified region reveals slight amorphous features at 100–200 cm⁻¹ and 430–500 cm⁻¹ and decreased crystalline characteristics.

Quantitative changes of the Raman signal at the waveguide region relative to the signal from unmodified silicon are listed in Table 1 as relative amplitude. Note that the amplitude of the main peak at 520 cm⁻¹ reduces to 58%. However, due to the fact that the main peak still exists at the center of the modified region, the silicon at the inner region of the waveguide basically consists of silicon with a disturbed crystalline structure.[25] Toward the outer region of the waveguide a continuous transition to the unperturbed crystalline structure can be seen. This is due to the Gaussian intensity profile at the laser focus yielding decreased
laser–matter interaction toward outer regions. These results are in good agreement with former works of Yater and Thompson, Liu et al., and Bonse et al.[20,26,27] They demonstrated the generation of polycrystalline and amorphous silicon at silicon surfaces affected by ns, ps, and fs laser pulses, respectively. Likewise Mori et al.[13] who study nanogratings in Si, see the same behavior in their Raman measurements. Peak-fitting of the crystalline contribution at 520 cm$^{-1}$ with a Lorentzian shape and a Gaussian shape for the amorphous contributions at 480 cm$^{-1}$ as well as for the grain boundaries contributions at 500 cm$^{-1}$ yield information on the degree of crystallinity.[28–30] Figure 3a–c) shows the values obtained for the fit of the Lorentzian shaped peak at 520 cm$^{-1}$ for the waveguide shown in Figure 1, 2. Apart from the already discussed reduction in amplitude (Figure 3b) a slight peak shift to a lower wave number of 518.9 cm$^{-1}$ (Figure 3a) as well as an increase of the width of the peak (Figure 3c) are clearly visible. The former is related to induced internal mechanical stress, where a movement to smaller wave numbers corresponds to tensile stress. A simple estimation of the average stress correlates a shift of 1 cm$^{-1}$ with respect to the stress free value (of about 520 cm$^{-1}$) to an uniaxial stress of 500 MPa or an in-plane biaxial stress of 250 MPa.[31] The peak broadening in general could have several origins as stress induced splitting of the three phonons of the 520 cm$^{-1}$ peak or large stress gradients in the tested volume. Additionally doping or impurities can cause a broadening due to new Raman peaks by their own vibrational modes. A third possible reason are extended crystal defects like grain boundaries, where also small grains in the nm range cause a broadening of the line width due to phonon confinement effects.[31] In our case, stress or extended grain boundaries are most likely.

Figure 3d) shows the crystalline fraction $F_C$ for each pixel, which can be calculated using the relation of the integrated areas $A$ of the peaks at 480 cm$^{-1}$, 500 cm$^{-1}$, and 520 cm$^{-1}$, respectively:[28]

$$F_C = \frac{A_{500} + A_{520}}{\alpha A_{480} + A_{500} + A_{520}},$$

(1)

where $\alpha = 0.8$ takes the higher phonon excitation cross-section of amorphous silicon into account.[28] Please note that the peak position was considered as free parameter during the fit.
procedure and $A$ is the overall sum over the fitted peak within the considered wave number range from 400 to 570 cm$^{-1}$. In the modified region $F_c$ decreases to 0.91. The amount of crystallinity is still high, as expected due to the distinct main peak. In conclusion the results of Raman peak fitting show at the modified region a reduction of the crystallinity, tensile stress, and a peak broadening, which could be caused by crystal defects.

In order to study the material modifications along the waveguide axis, waveguide arrays were densely inscribed using the same processing parameters as before. These samples were cleaved afterward yielding a plane, which goes through a few waveguides by chance and exposes these structures to the surface. Corresponding results can be seen in Figure 4.

In analogy to Figure 2 Raman mappings with a step size of 0.25 µm were generated. Figure 4 shows the 2D mappings of the amorphous feature at 430-500 cm$^{-1}$ (Figure 4a), the main c-Si peak at 520 cm$^{-1}$ (Figure 4b) and the crystalline feature at 930–1040 cm$^{-1}$ (Figure 4c). These measurements reveal modifications with an average diameter of 3–5 µm, as expected from the cross section measurements shown in Figure 2. The corresponding Raman spectra are consistent with the observations at the end facet, which confirms that the modifications extend into the volume of the material. The measured material changes are therefore not a surface effect or caused by the polishing of the end facet.

In order to understand the influence of these modifications, it is important to point out that the refractive index extremely depends on the induced crystal defects and involved residual strain. Amorphous silicon exhibits a significantly higher refractive index of 3.480 (at 1550 nm)\(^{32}\) compared to the mono-crystalline phase.
with a value of $3.47^{[31]}$ yielding a refractive index difference of $3 \times 10^{-3}$. The maximum calculated index change about $2 \times 10^{-3}$ (see Figure 1c,d) is in the same order of magnitude, which is consistent with the observation from Kühl et al.$^{[34]}$ In particular, strain affects the refractive index, however, an exact quantification is difficult and must be investigated in addition.$^{[35–38]}$

4. Conclusion

We investigated the origin of waveguiding in ultrashort pulse laser written structures in bulk silicon. This study was performed by Raman microscopy of the waveguide cross section and along the waveguide axis. The measurements reveal a permanently induced transition from mono-crystalline silicon to silicon with disturbed crystal structure within a cross section area of 3–5 µm. Moreover, a reduction of the crystalline fraction to $F_C = 91\%$ is observed. The decrease of $F_C$ reflects defects and potential dislocations. These modifications in combination with remaining strain are responsible for a positive refractive index change in the range of $10^{-3}$. This result is in good agreement with the refractive index distribution derived from mode-field measurements.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

laser inscription, raman microscopy, silicon waveguide, ultrashort pulses

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