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Effects of Photon Enhanced Lifetime and Form Anisotropy in Silicon Nanowire Arrays on Efficiency of Nonlinear-Optical Processes

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Abstract. Arrays of silicon nanowires (SiNWs) of about 100 nm in diameter formed by metal-assisted chemical etching of crystalline silicon (c-Si) substrates were studied regarding efficiency of optical interactions in them. Strong scattering in the SiNW arrays results in enhanced photon lifetime in them, which was evidenced by measurement of the cross-correlation function for femtosecond pulses scattered by the SiNW arrays. This effect results in an order of magnitude growth of efficiency such processes as spontaneous Raman scattering and third-harmonic (TH) generation compared to the signals for c-Si. Using (110) oriented c-Si substrate we managed to make well-oriented tilted SiNWs demonstrating strongly anisotropy of the TH and coherent anti-Stokes Raman scattering signals. In particular, efficiencies of these processes differ significantly when incident radiation propagates along or perpendicular to the SiNWs. The obtained results could be explained by variations of the local fields in SiNWs and scattering cross-section.

INTRODUCTION

Nowadays, arrays of silicon nanowires (SiNW) of about 100 nm in diameter attract more and more interest. Typically, SiNWs are aligned pillars with the length controlled by the formation procedure [1]. Among their optical properties we can mention extremely low reflection and high light absorption in the visible spectral region [2,3] resulting in promising industrial applications, e.g. for solar cell or sensor fabrication. In contrast, in near-infrared (NIR) spectral region thick SiNW layers demonstrate very high reflection. Besides, enhanced efficiency of many optical processes, including spontaneous Raman scattering (SpRS), third-harmonic (TH) generation, and infrared interband photoluminescence [2]. These effects are often connected with the light trapping in SiNW arrays caused by effective light scattering, which in its turn depends on length, orientation, and arrangement of the SiNWs. Thus, studying correlation of the SiNW structural parameters and efficiency of the optical processes, including nonlinear-optical ones seems to be very instructive for further applications of the SiNW arrays.
FIGURE 1. SEM images of SiNW arrays formed on (100) Si (a) and (110) Si (b) substrates.

EXPERIMENTAL

A series of SiNW arrays was fabricated on low boron-doped c-Si (100) polished wafers of (100) and (110) orientation via the MACE technique. The two-step procedure of SiNW formation was employed. First, silver nanoparticles were deposited on the c-Si surface during the immersion of Si wafer in the aqueous solution of 0.02 M AgNO₃ and 5 M HF in the volume ratio 1:1 for 1 minute. The second step was the etching of Si wafer covered by silver nanoparticles in solution of 5 M HF and 30% H₂O₂ in the volume ratio 10:1 for 5 minutes. Finally, silver nanoparticles were removed from the surface during rinsing the sample in HNO₃ for about 15 minutes. As a result, well-ordered parallel SiNWs of about 100 nm in diameter were formed (see Fig. 1). For (100) c-Si the SiNW are perpendicular to the surface, whereas for (110) substrate they are tilted to the surface at the angle of 45° with projection oriented along [1 10] direction.

To reveal effects of SiNW array structural parameters on efficiency of the optical processes we employed SpRS, TH generation, and coherent anti-Stokes Raman scattering (CARS), the dependence of the signals on the light polarization and thickness of the sample were found. These processes are sensitive to the light polarization and local field effects. The obtained results were complemented by measurements of cross-correlation function for the laser pulse and radiation scattered by SiNW arrays, which allows us to estimate photon lifetime in SiNW arrays. A quasi-cw Cr:forsterite laser (1250 nm, 80 fs, 150 mW, 80 MHz) was used for the TH generation and cross-correlation function measurement. The laser radiation was focused on the sample at the angle of incidence of 45° by a short-focus lens. For the orientation dependence detection the fundamental radiation polarization and analyzer were rotated simultaneously.

To obtain more detailed information on the SiNW arrays we carried out measurements of the SpRS in SiNW ensembles with the help of Fourier-transform IR spectrometer (Bruker IFS 66v/S) with a Raman unit FRA-106 FT. The experimental system employed in the CARS experiments was based on a diode-pumped Nd:YVO₄ master oscillator and a multipass amplifier (1064 nm, 10 ps, 2 μJ, 800 kHz). A part of the laser radiation was used for generation of broadband, red-shifted continuum radiation in a single-mode GeO₂-doped optical fiber, which was collimated and combined together through a dichroic beamsplitter with a time-delayed residual 1064 nm radiation. In the employed experimental setting, the broadband CARS signal was generated at the frequency 2ω₁ - ω₂, where ω₁ was the fundamental radiation frequency and ω₂ was the frequency of the continuum radiation. The CARS signal in the spectral region from 800 to 1030 nm was collected by a 30-mm lens directed to a spectrometer. Both fundamental and continuum channels were supplied with half-wave plates, whereas Glan-Taylor prism (analyzer) was used to select polarization of the generated CARS signal.

RESULTS AND DISCUSSIONS

Typical dependence of Raman and TH signals on the thickness of SiNW layer are shown in Fig. 2. Starting from different thicknesses, both dependences demonstrate sharp increase. For the TH signal the rise follows a decrease in the TH intensity for thinner layers (0.2–2 μm), while Raman signal grows monotonously from the thinnest SiNW layers and tends to saturation for the layer thickness above 5 μm. The difference is due to absorption of the TH radiation and coherent nature of the TH generation. It is worth noting the fact of significant variations of the
orientation dependences both for SpRS and TH signals with the SiNW layer thickness increase: the former one gets isotropic, whereas for the latter TH signal is anisotropic, with the orientation dependence varying with variation of the SiNW thickness.

The measurements of the cross-correlation functions revealed that for the SiNWs of the thickness above 4 μm the cross-correlation function has an expressed tail, indicating diffusion-like radiative transport in the SiNW array. In contrast, no significant variation of the cross correlation function was found for the arrays of less thickness. The typical photon lifetimes were found to be 0.5 and 1.4 ps for the SiNW arrays with thicknesses of 4.5 and 16 μm, respectively, which significantly exceeds the laser pulse duration. Note that the rise of the TH generation efficiency starts at the same thickness as the increase of the photon lifetime in the SiNWs. Such a correlation let us suppose that the latter one is responsible for the rise of the TH generation efficiency (Fig. 3).

In the case of the incident light propagation along the SiNWs the SpRS signal is highly depolarized, whereas for the incident light propagating perpendicular to SiNWs the difference of the \( p \)- and \( s \)-polarized SpRS signals reaches 25% and is weak polarized. In contrast to SpRS results, SiNW arrays exhibit pronounced polarization dependences of the CARS signal. The resonant CARS signal in SiNW ensemble is an order of magnitude less than in c-Si in the

![Figure 2](image1.png)

**FIGURE 2.** Raman (a) and TH (b) intensities at different thickness of SiNW layers. The intensity values are normalized to Raman signal from c-Si. The dashed line is eye guided. The inset in panel (a) shows a typical spectrum obtained under excitation at 1064 nm consisting of Raman peak and broad photoluminescent background.

![Figure 3](image2.png)

**FIGURE 3.** Auto-correlation function (a) and cross-correlation functions for the laser pulse and radiation scattered by the SiNW arrays with thicknesses 5 μm (c) and 16 μm (e) and powers of the corresponding signal (b, d, and f) vs. time.
FIGURE 4. (a) Polarization dependences of the resonant CARS signal for (110) c-Si and SiNW ensemble on continuum-radiation polarization. The upper line corresponds to c-Si wafer, the middle and bottom lines correspond to SiNWs at different geometries of the light incidence on them (along and perpendicular to SiNW, correspondingly). The rows 1 and 2 are for the CARS radiation polarized along [1 10] crystallographic direction, rows 3 and 4 for the CARS radiation polarized along [001] crystallographic direction. Polarization of the fundamental wavelength was along [1 10] (rows 1 and 3) and [001] (rows 2 and 4) crystallographic direction. Continuum-radiation polarization of 0° corresponds to [1 10] direction (perpendicular to the plane of incidence). (b) Orientation dependence of the third-harmonic signals for c-Si and SiNW array in cases of fundamental radiation incident along and perpendicular to the nanowires.

In conclusion, multiple scattering of near-infrared light in SiNW arrays leads to photon lifetime enhancement which is manifested by high reflection of thick silicon nanowire layers and substantial increase of the Raman scattering and TH generation efficiencies. Polarization dependences of the CARS resonant and TH signals strongly depends on to the incident wave propagation (perpendicular or parallel to the SiNW), with the light incidence perpendicular to the SiNWs being more effective than along the SiNWs. The obtained results could be explained by variations of in the local fields in SiNWs and scattering cross-section.

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