Anisotropy of third-harmonic and coherent anti-Stokes Raman scattering signals in silicon nanowire arrays

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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 high light absorption and enhanced efficiency of various optical processes, including spontaneous Raman scattering, 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 in them. However, despite this fact nonlinear-optical effects due to their pronounced dependence on the local field could be able to reveal anisotropy of the SiNW arrays.

In this paper we chose to study features of such nonlinear-optical processes as TH generation and coherent anti-Stokes Raman scattering (CARS) in the SiNW arrays of pronounced anisotropy formed by means of metal-assisted chemical etching process [1] at (110) crystalline silicon (c-Si) wafer. The SiNWs are strongly prolate parallelepipeds of about 100 nm in diameter tilted to the surface at the angle of 45◦ with projection oriented along (110) direction (Fig. 1). Due to effective light scattering in the SiNW arrays demonstrate no orientation effect in linear reflectance measurements.

The broadband CARS signal was generated at the frequency $2\omega_1$ - ω_2, where ω_1 and ω_2 were the frequencies of Nd:YVO4 laser (1064 nm, 10 ps) radiation and a continuum radiation generated in optical fiber, correspondingly. The TH generation was carried out with the help of Cr:forsterite laser (1250 nm, 80 fs). In all cases polarization dependences of the signals were measured.

In contrast to spontaneous Raman, the 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 case when pumping radiation propagates perpendicular to the SiNWs and the CARS signal is collected in the direction along SiNWs and two orders of magnitude less than in c-Si in another case. The TH signal for SiNW array exceeds one for c-Si in the case of the fundamental radiation incident perpendicular to the SiNWs, whereas in the case of incident wave propagation along the SiNWs it falls several time in comparison with c-Si (Fig. 2). Thus, found in experiments anisotropy of the nonlinear-optical signals evidences the sensitivity of these techniques to the orientation of SiNWs in their highly scattering arrays.

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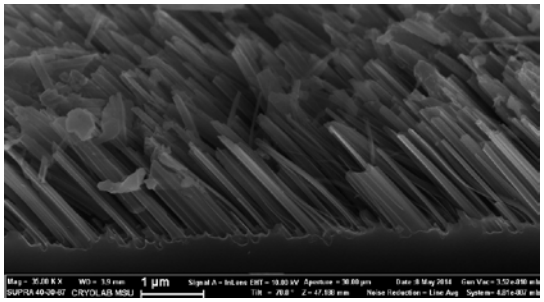

Fig. 1: The SiNW sample cross-section in (100) plane.

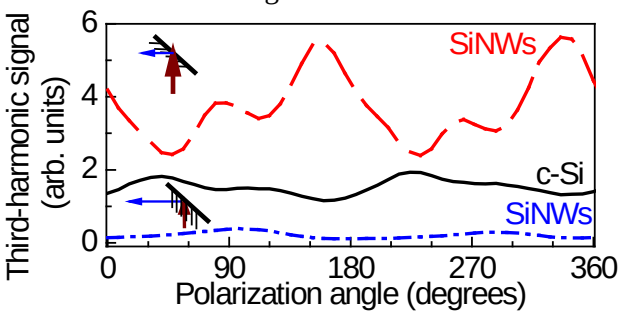

Fig. 2: Orientation dependence of the third-harmonic signals for c-Si and SiNW arrays for pump radiation incident along and perpendicular to the SiNWs.

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