Silicon nanoparticles for nonlinear frequency conversion of mid-IR radiation

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Abstract. Here a new way to use silicon nanoparticles for medium-infrared photonics is suggested. This contribution is focused on solving the fundamental problem of optical interaction between subwavelength silicon resonators and mid-IR laser radiation. In this spectral range the coefficient of two-photon absorption, which is the main mechanism for limiting the efficiency of nonlinear optical processes in silicon, is close to zero. The study provides answers to the fundamental questions about high-efficient nonlinear-optical transformations of four-wave mixing, and will serve as the basis for new nonlinear devices of integrated silicon nanophotonics.

1. Introduction
High-index nanoparticles are in the focus of many research groups specialized in photonics and nanophotonics. This interest appeared quite recently: it was observed that both electric and magnetic multipoles were excited in nanoparticles (spheres and disks) made out of high-index materials in the visible and near-IR spectral ranges [1-3], what is explained by Mie scattering theory [4].

The significant enhancement of the third-harmonic generation in a metasurface of closely packed silicon nanodisks comparing with non-structured area was detected [5] in the spectral vicinity of a magnetic dipolar resonance; it became the first work considering the study of nonlinear optical properties of all-dielectric nanostructures with optical magnetism. Moreover, optically isolated nanoparticles also reveal the effective up-conversion process at the magnetic dipolar Mie-type resonance [6] in contrast to the electric dipolar mode of a particle.

In order to increase the efficiency of nonlinear processes several nanostructure patterns have been studied including the combination of particles into oligomers, quadrumer [7], which leads to the excitation of collective optical Fano-type modes and 100-times enhancement of the third-harmonic-generation signal due to the redistribution of local electromagnetic field inside a silicon nanoparticle cluster.

The experimentally achieved nonlinear frequency conversion efficiency reaches values up to $10^{-5}$ in the consideration of the third-harmonic generation in silicon Mie-resonant particles; the main mechanism limiting the four-wave-mixing process efficiency is the two-photon absorption effect in silicon. The two-photon absorption coefficient is about 2.5 cm/GW in the near-IR spectral range for silicon [8, 9], whereas the coefficient becomes close to zero for a medium-infrared part of the spectrum, the multi-photon absorption parameters are also known for crystalline silicon [10].

The nonlinear optical process efficiency, particularly, the third-harmonic generation process in the case of silicon Mie-resonators, has not been studied for the mid-IR pump yet, as well as the excitation
of optical modes in silicon nanoparticles at triple pump frequencies, whilst the research of three-wave-mixing conversions in silicon nanoresonators with multipolar modes at the double pump frequency from the visible spectral range is conducted [11].

2. Idea
The two-photon absorption coefficient of silicon becomes close to zero for the wavelengths from 2.1 microns and longer, i.e. in the mid-IR spectral range. In order to keep the nanoscale of Mie-type resonator sizes it is suggested to consider nanoparticles supporting the excitation of electric and magnetic multipolar Mie-type optical modes at the frequencies of nonlinear optical response – at the triple pump frequencies.

3. Methods
Numerical simulations of the third-harmonic output from a silicon nanodisk on a glass substrate are performed by the finite-difference time-domain method with the help of the FDTD Solutions software by Lumerical Solutions, Inc. The material of the disk is considered as nonlinear one by adding the value of its third-order nonlinear susceptibility. Each spectral point is a simulation result for the specific central pump frequency of an external radiation source modeled by a plane wave pulse; this procedure also enables to take into account the dispersion of a silicon nonlinear susceptibility, its tensor nature is ignored. The optical signal at the vicinity of a third-harmonic generation wavelength is detected by a two-dimensional monitor and, then, integrated as a function of these two spatial coordinates and frequency.

The sizes of considered silicon disks provide the excitation of multipolar resonances at the triple pump frequencies which correspond to the spectral range from 650 to 800 nm.

4. Discussion
Performed calculations reveal the strong enhancement of the third-harmonic generation signal from silicon nanoparticles at the vicinity of the magnetic dipolar resonance of a silicon nanodisk when the optical mode is excited at the triple pump frequency. The maximum value of normalized nonlinear signal is observed to be shifted in the blue area of the spectrum with respect to the Mie-type resonance due to dispersion dependencies of optical material parameters taken into account. These spectral features of the nonlinear response qualitatively coincide with the experimental findings reported previously for a silicon disk metasurface with spectrally overlapped electric and magnetic dipolar modes [12] and explain them by the model chosen for the numerical simulations and described above.

The proposed approach provides the resonant efficiency enhancement of the four-wave-mixing process in silicon nanoparticles excited by the mid-IR radiation as well as overcomes the fundamental conversion efficiency limit due to the two-photon absorption effect in silicon. These findings open a new avenue for novel nonlinear photonic devices.

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