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Investigation of second harmonic generation in spherical mesoporous Si/SiO_2 nanoparticles on gold

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Abstract. In this work, experimental and numerical investigation of second harmonic generation (SHG) in mesoporous Si/SiO_2 nanoparticles has been performed. Experimental results are well-described by simulations. Spectral analysis reveals that SHG efficiency maxima correlate with Mie resonances of Si/SiO_2 nanoparticles. Tuning the diameter of the structures the maximum SHG efficiency for required wavelength can be achieved. The nonlinear optical susceptibility of the studied nanoparticles attains values on the order of 1.59×10^{-14} m^2/V, which exceeds that of bulk silicon. Spherical mesoporous Si/SiO_2 nanoparticles demonstrate effective second harmonic generation with simple, low-cost fabrication, making them promising candidates as tunable frequency converters for integrated nanophotonic circuits.

Keywords: second harmonic generation, silicon, spherical nanoparticles, Mie resonances, gold, mesoporous

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Исследование генерации второй гармоники в сферических мезопористых наночастицах Si/SiO_2 на золоте

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Аннотация. В данной работе проведено экспериментальное и численное исследование генерации второй гармоники (ГВГ) в мезопористых наночастицах Si/SiO_2. Спектральный анализ показывает, что максимумы эффективности ГВГ коррелируют с резонансами Ми наночастиц Si/SiO_2. Путем настройки диаметра структур можно достичь максимальной эффективности ГВГ для требуемой длины волны. Нелинейная оптическая восприимчивость исследуемых наночастиц достигает значений порядка $1,59\times10^{-14}$ м^2/В, что превышает значения для объемного кремния. Сферические мезопористые наночастицы Si/SiO_2 демонстрируют эффективную генерацию второй гармоники при простом и недорогом изготовлении, что делает их перспективной альтернативой в качестве перестраиваемых преобразователей частоты для интегральных нанофотонных схем.

Ключевые слова: генерация второй гармоники, кремний, сферические наночастицы, Ми резонансы, золото, мезопористый

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Introduction

Second-order nonlinear optical effects are essential for coherent light sources, optical amplifiers, and nanophotonic devices, particularly for converting infrared to visible light via second harmonic generation (SHG). While nonlinear crystals like KDP and $LiNbO_3$ [1, 2] and III-V semiconductors [3] offer efficient frequency conversion, their high cost and limited silicon compatibility restrict integration into modern optical systems.

Silicon platforms provide cost-effective fabrication of diverse structures but suffer from poor second-order nonlinear efficiency due to silicon's symmetry of the crystal lattice, which eliminates bulk SHG. However, SHG can be allowed through symmetry breaking at crystal boundaries [4] and higher-order interactions. Since SHG depends on morphology and surface properties, approaches include porous silicon and silicon nanoparticles/nanowires with favorable second-order characteristics [5]. Additional enhancement approaches include optical cavities for electromagnetic field enhancement, which are formed by dielectric or plasmonic materials.

Previous work, devoted to the study of the mesoporous Si/SiO_2 nanoparticles [6], represents experimental results for the array of the nanoparticles. In this work, investigation of the second harmonic generation in single mesoporous nanoparticles of different diameters was held numerically and experimentally. Nanoparticles are a framework of silicon oxide (SiO_2) filled with nanocrystalline silicon.

Materials and Methods

The mesoporous Si/SiO_2 nanoparticles have complex structure that is challenging to construct and calculate numerically. However, when material crystallites are much smaller than the incident wavelength, the Bruggeman effective medium approximation can describe the optical properties of these composite materials. Numerical calculations were performed using COMSOL Multiphysics for spherical mesoporous Si/SiO_2 nanoparticles of different diameters. The model used Si/SiO_2 particles laying on gold substrate and surrounded by air shell, with incident wavelengths of 840–1000 nm. Refractive indices and extinction coefficients for silicon and silicon oxide taken from references [7] and [8], respectively.

Nonlinear polarization, which determines the SHG in the studied Si/SiO_2 nanoparticles on gold is considered as follows:

$$\vec{P}^{NL} = \vec{P}^{surf} + \vec{P}^{cryst},$$

$$\vec{P}^{surf} = \varepsilon_0 \delta(z)\left[\hat{n}\left(\chi^{(2)}_{\perp\perp\perp}\left(E_n^{(\omega)}\right)^2 + \chi^{(2)}_{\perp\|\|}\left(E_\tau^{(\omega)}\right)^2\right) + 2\hat{\tau}\chi^{(2)}_{\|\perp\|} E_n^{(\omega)} E_\tau^{(\omega)}\right],$$

$$\vec{P}^{cryst} = \frac{1}{V_{cryst}} \int\limits_{V_{cryst}} \varepsilon_0 \delta(z) \chi^{surf}\left(\vec{E}_0 + \hat{D}\vec{E}\cdot\vec{r}\right)\left(\vec{E}_0 + \hat{D}\vec{E}\cdot\vec{r}\right) d^3\vec{r}.$$

There are 2 possible mechanisms of SHG: surface SHG $\vec{P}^{surf}$, determined by violations of translational symmetry of the crystal lattice and crystallites SHG $\vec{P}^{cryst}$. $\chi^{(2)}_{\perp\perp\perp}$, $\chi^{(2)}_{\perp\|\|}$, $\chi^{(2)}_{\|\perp\|}$ – components of the surface tensor of nonlinear optical susceptibility.

Second harmonic measurements were performed using a Zeiss LSM-980 confocal scanning laser microscope with a Coherent Discovery-NX acousto-optic modulator. The sample was excited by 150 fs femtosecond pulses at 80 MHz repetition rate with maximum power of ~25 MW (0.3 nJ per pulse). Signals were detected in reflection geometry.

Results and Discussion

Experimental and numerical results of the investigation are represented on Fig. 1.

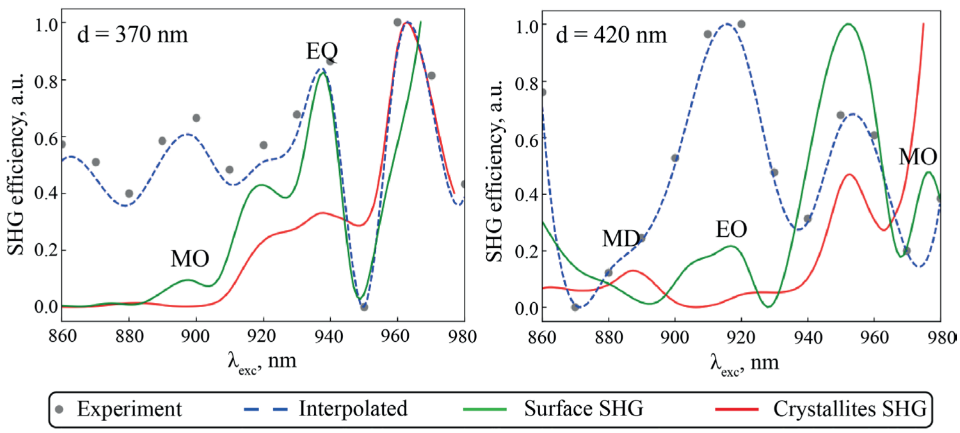

Fig. 1. SHG efficiency spectra (λ_{exc} – excitation wavelength) for mesoporous Si/SiO_2 nanoparticles with diameters d = 370 nm and 420 nm

Experimental spectra has different maxima, representing enhancement in nonlinear response. However, the nature of these peaks can be figured out using numerical calculations. Thus, the results of the numerical calculations, related to different SHG mechanisms, were plotted in the experimental graphs. In Fig. 1, one can see two main contributions to SHG from mesoporous Si/SiO_2 nanoparticles: external surface contribution and nonlinear response from the crystallites edges. Both this mechanisms shows some maxima that can be related to different Mie and hybrid resonances formed by nanoparticle and gold substrate. The calculated nonlinear optical susceptibility of the considered nanoparticles under resonance conditions can reach values on the order of 1.59×10^{-14} m^2/V, which exceeds that of bulk silicon [9].

Comparing experimental and numerical results one can talk about the relationship of the maxima of the spectra with the resonances at different wavelengths. Both mechanisms of SHG in nanoparticles under investigation contribute to experimental nonlinear response. All peaks obtained experimentally are described by calculated curves adjusted for their intensity. This correction may be related to the inaccuracy of modeling in relation to experimental conditions. Nevertheless, we can talk about a good agreement between experiment and theory.

Conclusion

In this work, the results of a numerical and experimental study of the second harmonic response from single mesoporous Si/SiO_2 nanoparticles are considered. SHG in such strictures is mainly described by two mechanisms: external surface SHG and crystallites edges SHG.

It was found that both SHG mechanisms contribute to nonlinear response in the nanoparticles, which manifests itself as an increased efficiency of second harmonic generation at resonant wavelengths.

Thus, the possibility of effective generation of the second harmonic in spherical mesoporous Si/SiO_2 nanoparticles is shown. The simplicity, as well as the low cost of their manufacture, makes it possible to propose these structures as efficient tunable frequency converters for nanophotonic circuits.

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