Semiconductors, 2024, Vol. 58, No. 9

UDC 621.315.592

# Numerical modeling of the second harmonic generation in Si nanoparticles

© A.S. Funtikova[1], A.M. Mozharov[2], V.A. Mastalieva[2], V.V. Neplokh[2], I.S. Mukhin[1,2], V.V. Fedorov[1]

[1] Alferov Federal State Budgetary Institution of Higher Education and Science
Saint Petersburg National Research Academic University of the Russian Academy of Sciences,
194021 St. Petersburg, Russia
[2] Peter the Great Saint-Petersburg Polytechnic University,
195251 St. Petersburg, Russia

E-mail: n.fn@mail.ru

*Received May 2, 2024*
*Revised July 13, 2024*
*Accepted October 30, 2024*

In this work, the effect of intrinsic optical resonances of spherical Si nanoparticles on the second harmonic generation process was studied numerically. An increase in the efficiency of the second harmonic generation process was found due to the resonant amplification of the incident electric field or the resonant phasing of the second harmonic near field due to the Mie resonances..

**Keywords:** second harmonic, silicon, spherical nanoparticles, Mie resonances.

DOI: 10.61011/SC.2024.09.59909.6236A

## 1. Introduction

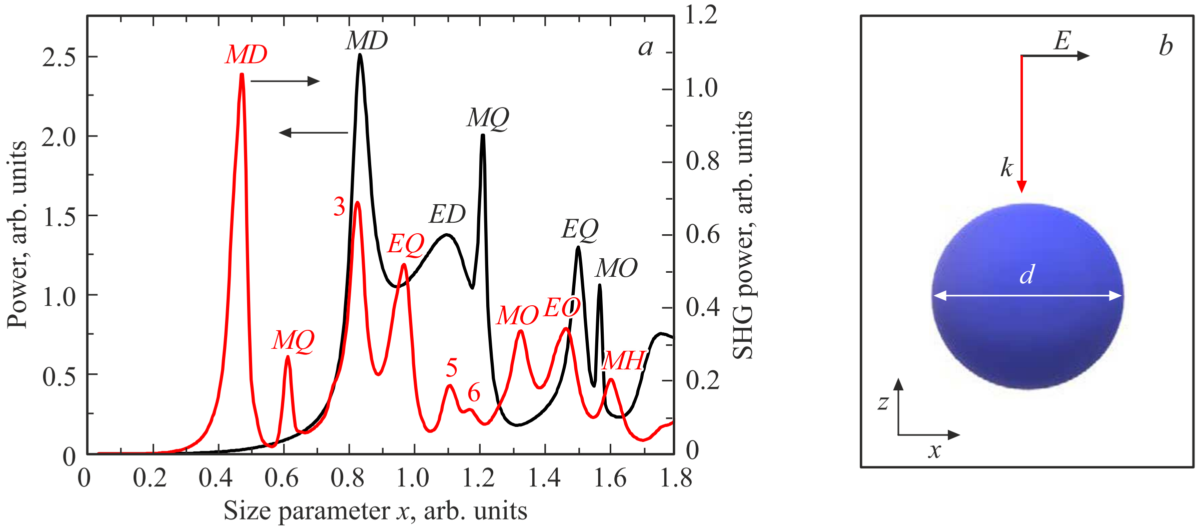
Second order nonlinear optical phenomena are commonly used for the creation of coherent light sources, frequency converters, optical amplifiers and other devices. $A^{III}B^{V}$ compounds are the most common semiconductor materials for these applications. Devices with high efficiency of doubling the frequency of incident radiation can be created based on these materials due to high values of nonlinear optical susceptibility [1,2]. In turn, high cost of their synthesis, as well as the limitation of their integration into traditional optoelectronic devices and integrated photonics elements based on silicon are serious disadvantages of structures based on $A^{III}B^{V}$ compounds. The silicon platform allows creating a variety of two-dimensional and three-dimensional structures. The low conversion efficiency of incident optical power in the framework of nonlinear processes is the main problem of the active development of nonlinear photonics on silicon. It is known that the second-harmonic generation (SHG) is impossible in a bulk material with a diamond-type crystal structure, which is attributable to the presence of a center of inversion of the crystal lattice. Despite this, there are several SHG mechanisms in centrosymmetric media both on the surface and in the structure itself, determined by the possibility of lowering the symmetry of the lattice or the process itself. If a decrease in symmetry inside the crystal is achieved by an inhomogeneity of the electric field, then the crystal surface itself has a reduced lattice symmetry due to a interruption of the translational symmetry [3,4]. This mechanism is implemented at short distances from the surface, on the order of several atomic layers. The deformation of the electron density through the polarization of the material in a constant or slowly changing electric field of spatial charge (EFISH — electric-field-induced second harmonic generation) is another mechanism of symmetry reduction [5]. The advantage of this mechanism is the ability to control the distribution of spatial charge fields, which opens up space for improving the efficiency of the SHG process in silicon and silicon-metal nanostructures [6]. Since the surface charge is the main source of electric fields in the crystal, the EFISH process is strongly influenced by the distribution of the incident field, which, in particular, depends on the presence of optical resonances in the system, which requires to study the effect of such resonances on the EFISH process efficiency, which is the subject of this paper.

## 2. Calculation procedure

In the general case, the SHG is determined by the appearance of a quadratic term in the decomposition of the polarization of the medium $P$ according to the degrees of intensity of the incident electric field [7]. In the case of the EFISH field mechanism, it can be represented as [5]

$$P = \hat{\chi}^{(3)}\mathbf{E}^{DC}\mathbf{E}^{\omega}\mathbf{E}^{\omega} = \hat{\chi}^{(2)}_{EFISH}\mathbf{E}^{\omega}\mathbf{E}^{\omega}, \qquad (1)$$

where $\mathbf{E}^{DC}$, $\mathbf{E}^{\omega}$ — electric field strength vectors of the space charge and incident light, respectively, $\chi^{(3)}$ — tensor of 3rd order nonlinear optical susceptibility, $\chi^{(2)}_{EFISH}$ — the tensor of nonlinear optical susceptibility of the 2nd order, which determines the generation of the second harmonic by the EFISH mechanism. It is worth noting separately that the field $E_{DC}$ can be formed by free optically induced charges in addition to uncompensated bound surface charges, which also have a significant effect on the near-surface field of the crystal [6].




*a* — graph of the dependence of the efficiency of light scattering and second harmonic generation on the relative diameter of the particle $x = \pi d/\lambda$. The end-to-end numbering of peaks on the dependence is presented, and some types of resonances are also indicated: $E/MD$ — electric/magnetic dipole, $Q$ — quadrupole, $O$ — octupole, $H$ — hexadecapole; *b* — geometric configuration of the system, $k$ — wave vector of incident light, $d$ — particle diameter, $E$ — electric field strength of incident light.

Numerical simulation was performed using the COMSOL Multiphysics package. Spherical Si-particles with diameters $d = 10-550$ nm, wavelength of incident light $\lambda = 920$ nm were considered. Estimates of the surface electric field [6] show that an intensity of $E^{DC} \sim 10^8$ V/m can be achieved in the Si-particle system with SCR width of $\sim 4$ nm, which allows estimating the value of the effective nonlinear optical susceptibility of $\chi^{(2)}_{EFISH} \sim 17$ pm/V, which is a good indicator for their application [8]. The constant electric field was set by a linearly increasing function near the surface of the particles with a maximum value at the boundary of Si and decreasing to zero at a depth of 4 nm. A plane linearly polarized electromagnetic wave was used as the radiation source. The spectral dependences of the refractive and extinction indices for Si were taken from [9], the values of the nonlinear optical susceptibility of 3rd order $\chi^{(3)}_{1111}$ were taken from [10].

## 3. Results and discussion

The results obtained are shown in the figure as dependences of the efficiency of elastic scattering of the incoming radiation and generation of the SHG signal on the reduced sizes of spherical Si-particles ($x = \pi d/\lambda$). This value was calculated using the following formula:

$$P = \frac{4 \iint S_n dS}{I_0 \pi d^2}, \tag{2}$$

where $S_n$ — the surface-normal component of the Poynting vector, $S$ — the area of the outer surface of the air layer surrounding the particle, $I_0$ — the intensity of incident light, $d$ — particle diameter.

Rayleigh scattering is observed in both dependencies for small $x$, which, as the particle size increases, is replaced by peaks characterictic of resonances during Mie scattering describing various multipole states of the electromagnetic field. For light scattered by a dielectric particle, the first maximum on the graph, according to Mie theory, is associated with a magnetic dipole $MD$, the second peak is associated with an electric dipole $ED$, the third peak is associated with a magnetic quadrupole $MQ$, etc. [11,12]. Similar peaks are also observed in the SHG spectrum at half the relative sizes: these are the natural resonances of the system (particles of the same absolute size $d$) at a wavelength of $\lambda/2$. The appearance of peaks at $\pi d/\lambda = 0.81$, 1.11 and 1.17 on the SHG efficiency curve is not related to the presence of resonances at the wavelength of the second harmonic, but is explained by the influence of $MD$, $ED$ and $MQ$ resonances of the incident field, respectively. Thus, the amplification of the SHG signal can occur both due to the resonant localization of light associated with the occurrence of Mie resonances in the nanoparticle at the pump wavelength, and due to resonances at the wavelength of the second harmonic.

It is noticeable that there is no corresponding maximum on the graph of the second harmonic for the peak of the electric quadrupole $EQ$ on the graph of the scattered field (see figure). This may be due to the fusion of the peak caused by the resonance of the incident field and the intrinsic resonance $EO$ at twice the frequency and too little gain of the SHG due to this resonance. Also, there is no maximum for the SHG signal corresponding to the electric dipole $ED$ in the Mie theory. This feature is related to the type of tensor $\chi^{(2)}_{EFISH}$, which includes the symmetry of the crystal lattice and structure, as well as the direction of polarization of the incident wave [13].

## 4. Conclusion

The results obtained allow making a conclusion that it is possible to enhance the intensity of the second harmonic signal using two mechanisms: resonant localization of light in the nanoparticle at the pump wavelength and Mie resonances at the second harmonic wavelength. Thus, it is possible to talk about increasing the efficiency of SHG in nanoparticles by using spheres whose diameters correspond to the appearance of electric or magnetic Mie resonances for the wavelength of incident light or the second harmonic. These results can be used to create efficient frequency converters, coherent light sources, and other nonlinear optical devices.

### Funding

The work was carried out with the support of the Ministry of Science and Higher Education (State assignment grant FSEG-2024-0017). V.V. Neplokh thanks the Russian Science Foundation for financial support of the study of nonlinear optical processes in Si-particles (agreement 23-79-00018).

### Conflict of interest

The authors declare that they have no conflict of interest.

## References

[1] A.P. Anthur, H. Zhang, Y. Akimov, J. Rong Ong, D. Kalashnikov, A.I. Kuznetsov, L. Krivitsky. Opt. Express, **29** (7), 1 (2021). DOI: 10.1364/oe.409758
[2] K. Rivoire, S. Buckley, F. Hatami, J. Vučković. Appl. Phys. Lett., **98** (26), 263113-1 (2011). DOI: 10.1063/1.3607288
[3] O.A. Accipetrov, V.O. Bessonov, T.V. Dolgova, A.I. Maydykovsky. JETP Lett., **90** (11), 813 (2009). (in Russian). DOI: 10.1134/S0021364009230064
[4] B. Huo, X. Wang, S. Chang, M. Zeng. J. Opt. Soc. Am. B, **29** (7), 1631 (2012). DOI: 10.1364/JOSAB.29.001631
[5] A. Widhalm, C. Golla, N. Weber, P. Mackwitz, A. Zrenner, C. Meier. Opt. Express, **30** (4), 4867 (2022). DOI: 10.1364/oe.443489
[6] Y. Sun, A. Larin, A. Mozharov, E. Ageev, O. Pashina, F. Komissarenko, I. Mukhin, M. Petrov, S. Makarov, P. Belov, D. Zuev. Light: Sci. Appl., **12** (1), 237-1 (2023). DOI: 10.1038/s41377-023-01262-8
[7] V.N. Kapshai, A.I. Tolkachev, A.A. Shamyna. Opt. i spektr., **129** (12), 1537 (2021). (in Russian). DOI: 10.21883/OS.2021.12.51742.2385-21
[8] C.K.N. Patel. Phys. Rev. Lett., **16**, 613 (1966). DOI: 10.1103/PhysRevLett.16.613
[9] M.A. Green. Solar Energy Mater. Solar Cells, **92** (11), 1305 (2008). DOI: 10.1016/j.solmat.2008.06.009
[10] N.K. Hon, R. Soref, B. Jalali. J. Appl. Phys., **110** (1), 011301-1 (2011). DOI: 10.1063/1.3592270
[11] A.B. Evlyukhin, S.M. Novikov, U. Zywietz, R.L. Eriksen, C. Reinhardt, S.I. Bozhevolnyi, B.N. Chichkov. Nano Lett., **12** (7), 3749 (2012). DOI: 10.1021/nl301594s
[12] D. Timbrell, J.W. You, Y.S. Kivshar, N.C. Panoiu. Sci. Rep., **8** (1), 3586 (2018). DOI: 10.1038/s41598-018-21850-8
[13] K. Frizyuk, I. Volkovskaya, D. Smirnova, A. Poddubny, M. Petrov. Phys. Rev. B, **99**, 075425 (2019). DOI: 10.1103/PhysRevB.99.075425

*Translated by A.Akhtyamov*