Modeling of second harmonic generation by a hybrid bi-resonant nanoantenna

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Abstract. Dielectric materials with high nonlinear susceptibilities are basic elements in biophotonics applications, such as biosensing or bioimaging [1,2]. Their nonlinear response can be significantly improved by plasmonic resonances of metal nanoantennas. In our work we combine a hybrid metal-dielectric nanoantenna, consisting of metal part having resonance at incident wavelength, and a nonlinear dielectric nanoantenna which has resonance at second harmonic providing enhancement of nonlinear signal generation. We performed numerical simulation of hybrid nanoantenna and demonstrated that the generation of second harmonic is 50 times more effective compared to bare dielectric nanoparticle.

1. Introduction

Second harmonic generation (SHG) is widely used in biological and medical science for imaging of biological objects (e. g. cells, molecules). For instance, SHG microscopy allows to observe SHG from single molecules, labeled with dyes that have large nonlinear response [1,2]. However, efficiency of SHG by nanoscale nonlinear crystals is very weak [3]. One of the solutions to overcome poor frequency conversion efficiency is to place nonlinear particle at a hot spot of plasmonic nanoantenna, which possesses surface plasmon resonances [4]. Thus, adjusting the hybrid nanoantenna to be resonant for both: the incident field and SHG, one can expect high SHG enhancement. In our work, we used $BaTiO_3$ as a nonlinear particle and combined it with two plasmonic gold nanoparticles (Figure 1) to achieve more effective SHG with respect to bare $BaTiO_3$.

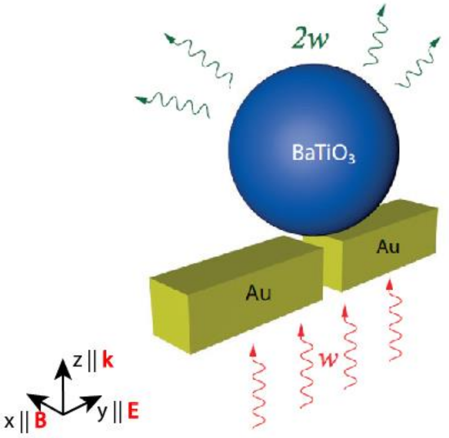

Figure 1. The metal-dielectric nanoantenna under consideration.

2. Simulation

We aimed at effective SHG in the visible spectrum, particularly at 600 nm. Hence, we adjusted $BaTiO_3$ particle to have a Mie resonance at exactly 600 nm wavelength, and the whole hybrid nanoantenna to have a plasmon resonance at 1200 nm. To analyze resonant properties of metal-dielectric nanoantennas we used numerical simulation in COMSOL Multiphysics environment, based of finite-element method. Investigated system was successively exposed to a plane wave with wavelengths between 800 nm and 1500 nm. The plane wave was propagating along positive direction of z-axis (Figure 1); **E** was oscillating along y-axis. To find electrical field distribution a Helmholtz equation was numerically solved. The values of gold [5] and $BaTiO_3$ [6] permittivity were taken from literature.

For simulation of SHG, we introduced quadratic polarization vector $\mathbf{P}^{(2)}$ of $BaTiO_3$, which is defined by quadratic electric susceptibility $\chi^{(2)}$ as $P_i=\chi_{ijk}^{(2)}E_jE_k$, and can be written in form of rectangular tensor of second order:

$$\begin{pmatrix} 0 & 0 & 0 & 0 & \chi_{15} & 0 \\ 0 & 0 & 0 & \chi_{15} & 0 & 0 \\ \chi_{31} & \chi_{31} & \chi_{33} & 0 & 0 & 0 \end{pmatrix}$$

Values of χ_{31}, χ_{33}, χ_{15} are taken from the experimental data [7].

3. Results and discussion

After simulation of scattering on metal-dielectric nanoantenna, we found a configuration with plasmon resonance at 1200 nm and Mie resonance of nonlinear particle at 600 nm. The geometry of nanoantenna is shown in Figure 2(a-c). We calculated SHG cross section of this hybrid nanoantenna and compared it with SHG cross section of a bare $BaTiO_3$.We also calculated SHG for a configuration without matching between plasmon resonance wavelength and double Mie resonance wavelength (Figure 2(b)) and for a configuration without a hotspot (Figure 2(c)). We demonstrated that both, resonant wavelengths matching and hotspot existence, result into additional SHG enhancement (Figure 2(d)). The calculated SHG enhancement with hybrid nanoantenna was up to 50 times.

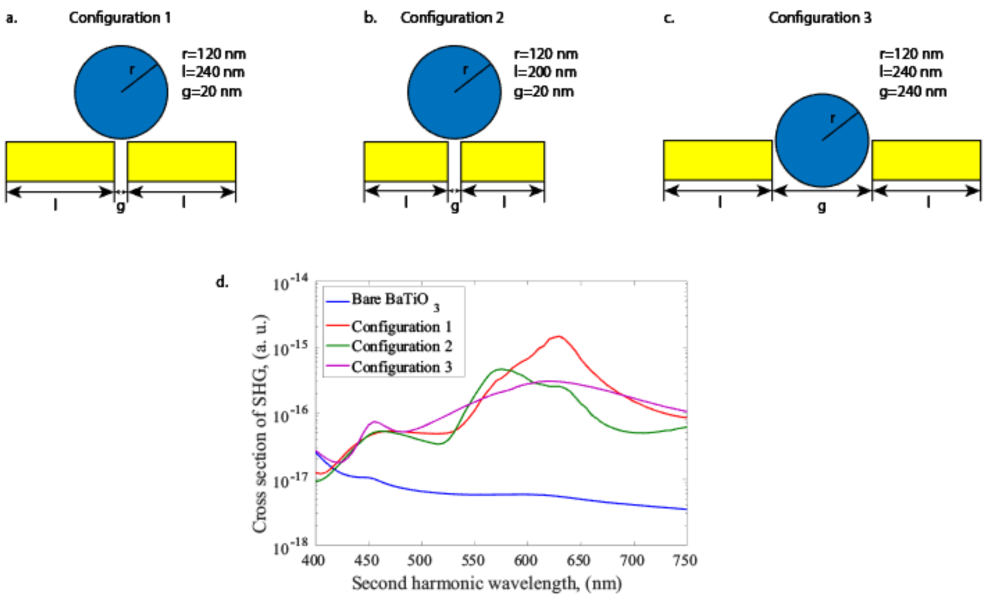

Figure 2(a, b, c, d). (a) Configuration 1 (with both, a hotspot and resonant wavelengths matching); **(b)** Configuration 2 (only with a hotspot); **(c)** Configuration 3 (with no hotspot or resonant wavelengths matching); **(d)** SHG for all 3 configurations and bare $BaTiO_3$ particle.

References

[1] G. Peleg, A. Lewis, M. Linial and L. M. Loew 1999 *Proc.Natl.Acad.Sci.USA* **96** 6700
[2] P. J. Campagnola, M. Wei, A. Lewis and L. M. Loew 1999 *Biophys.J.* **77** 3341
[3] Staedler D, Magouroux T, Hadji R, Joulaud C, Extermann J et al. 2012 *ACSNano* **6** 2542
[4] Linnenbank, H.; Grynko, Y.; Forstner, J.; Linden and S. Light 2016¨ *Sci.Appl.* **5** 16013
[5] S. Babar and J. H. Weaver 2015 *OSAPublishing* **54** 477
[6] S.H.Wemple; M.Didomenico anf Jr.I.Camlibel 1968 *J.Phys.Chem.Solids* **29** 1797
[7] Weber M. J. 2002 *HandbookofOpticalMaterials* (CRC press) **19**